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Silicic Magma Genesis in Basalt-dominated Oceanic Settings

Examples from Iceland and the Canary Islands

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Abstract
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The origin of silicic magma in basalt-dominated oceanic settings is fundamental to our understanding of magmatic processes and formation of the earliest continental crust. Particularly significant is magma-crust interaction that can modify the composition of magma and the dynamics of volcanism. This thesis investigates silicic magma genesis on different scales in two ocean island settings. First, volcanic products from a series of voluminous Neogene silicic centres in northeast Iceland are investigated using rock and mineral geochemistry, U-Pb geochronology, and oxygen isotope analysis. Second, interfacial processes of magma-crust interaction are investigated using geochemistry and 3D X-ray computed microtomography on crustal xenoliths from the 2011-12 El Hierro eruption, Canary Islands.

The results from northeast Iceland constrain a rapid outburst of silicic magmatism driven by a flare of the Iceland plume and/or by formation of a new rift zone, causing large volume injection of basaltic magma into hydrated basaltic crust. This promoted crustal recycling by partial melting of the hydrothermally altered Icelandic crust, thereby producing mixed-origin silicic melt pockets that reflect the heterogeneous nature of the crustal protolith with respect to oxygen isotopes. In particular, a previously unrecognised high-$\delta^{18}O$ end-member on Iceland was documented, which implies potentially complex multi-component assimilation histories for magmas ascending through the Icelandic crust. Common geochemical traits between Icelandic and Hadean zircon populations strengthen the concept of Iceland as an analogue for early Earth, implying that crustal recycling in emergent rifts was pivotal in generating Earth's earliest continental silicic crust.

Crustal xenoliths from the El Hierro 2011-2012 eruption underline the role of partial melting and assimilation of pre-island sedimentary layers in the early shield-building phase of ocean islands. This phenomenon may contribute to the formation of evolved magmas, and importantly, the release of volatiles from the xenoliths may be sufficient to increase the volatile load of the magma and temporarily alter the character and intensity of an eruption.

This thesis sheds new light on the generation of silicic magma in basalt-dominated oceanic settings and emphasises the relevance of magma-crust interaction for magma evolution, silicic crust formation, and eruption style from early Earth to present.

Keywords: Silicic magmatism, Iceland, magma-crust interaction, proto-continental crust, early Earth, zircon geochronology and geochemistry, oxygen isotopes, 2011-2012 El Hierro eruption, crustal xenoliths, 3D X-ray μ-CT, volatiles

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Believe you can and you're halfway there.

Theodore Roosevelt

List of Papers

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

I **Berg, S. E.**, Troll, V. R., Burchardt, S., Riishuus, M. S., Krumbholz, M., Gústafsson, L. E. (2014). Iceland's best kept secret. *Geology Today*, 30(2), 54–60.

II **Berg, S. E.**, Troll, V. R., Burchardt, S., Riishuus, M. S., Deegan, F. M., Harris, C., Whitehouse, M. J., Krumbholz, M., Gústafsson, L. E. (in prep.). Rapid generation of high-silica magmas in basalt-dominated rift settings. Manuscript intended for *Scientific Reports*.

III **Berg, S. E.**, Troll, V. R., Deegan, F. M., Ellis, B. S., Burchardt, S., Riishuus, M. S., Meade, F. C. (in prep.). Zircons from northeast Iceland analogous to those from early Earth. Manuscript intended for *Geology*.

IV **Berg, S. E.**, Troll, V. R., Harris, C., Riishuus, M. S., Deegan, F. M., Burchardt, S. (in prep.). Origin of high whole-rock $\delta^{18}O$ values in rhyolites from northeast Iceland. Manuscript intended for *Mineralogical Magazine*.

V Troll, V. R., Klügel, A., Longpré, M.-A., Burchardt, S., Deegan, F. M., Carracedo, J. C., Wiesmaier, S., Kueppers, U., Dahren, B., Blythe, L. S., Hansteen, T., Freda, C., Budd, D. A., Jolis, E. M., Jonsson, E., Meade, F., **Berg, S. E.**, Mancini, L., and Polacci, M. (2012). Floating stones off El Hierro, Canary Islands: xenoliths of pre-island sedimentary origin in the early products of the October 2011 eruption. *Solid Earth* 3, 97–11.

VI **Berg, S. E.**, Troll, V. R., Burchardt, S., Deegan, F. M., Mancini, L., Polacci, M., Carracedo, J. C., Soler, V., Zaczek, K., Krumbholz, M., Brun, F., Arzilli, F. (in prep.). Heterogeneous vesiculation of 2011 El Hierro xeno-pumice revealed by synchrotron µ-CT. Manuscript intended for *Bulletin of Volcanology*.

VII **Berg, S. E.**, Kim-Andersson, A. (2014). Hur bildades kontinenterna? *Geologiskt forum* 84, 12–13. A popular science article.

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Personal Contributions

The manuscripts included in this thesis are the result of multi-author team efforts. My individual contributions to each of them are as follows:

Paper I: My contribution was about 65 % of the total effort. I wrote the initial manuscript draft and prepared the figures.

Paper II: My contribution was about 60 %. I carried out fieldwork and sampling together with co-authors, undertook all sample preparations and characterisations, and subsequent SIMS analyses. I performed data processing and geochemical modelling, prepared the figures and wrote the initial draft of the manuscript in collaboration with the co-authors.

Paper III: My contribution was approximately 70 %. I formulated the research topic, organised and carried out fieldwork and sampling, and undertook sample preparations. I characterised the samples, performed geochemical analyses and participated in data reduction. I completed the geochemical data modelling, prepared the figures and wrote the manuscript with contributions from all co-authors.

Paper IV: My contribution was around 65 %. I carried out fieldwork and sampling together with co-authors, and undertook sample preparations and geochemical modelling. I organised data discussions, prepared the figures and wrote the initial draft of the manuscript that evolved with significant input from the author team.

Paper V: My contribution was ~5 %. I was involved in sample preparations and participated in data discussions.

Paper VI: My contribution was about 60 %. I performed 3D reconstructions, modelling and quantitative analyses of samples that were collected for paper V, with assistance provided by the Elettra staff. I interpreted the obtained dataset, organised data discussions, prepared the manuscript figures and wrote the initial draft with support from the entire author team.

Paper VII: I wrote the initial draft of the article that was complemented with an interview. I provided photos and proof read the editor's final version of the article.

Contents

Abbreviations

AFC	Assimilation and Fractional Crystallisation
CCD	Charge Coupled Device
CL	Cathodoluminescence
EH	El Hierro
EMPA	Electron Microprobe Analysis
Ga	Billion years ago
HP-HT	High Pressure-High Temperature
LA-ICP-MS	Laser Ablation Inductively Coupled Plasma Mass Spectrometry
LOD	Limits of Detection
Ma	Million years ago
MAR	Mid-Atlantic Ridge
MORB	Mid Ocean Ridge Basalt
MSWD	Mean Square Weighted Deviation
NAIP	North Atlantic Igneous Province
P	Pressure
REE	Rare Earth Element
SEM	Scanning Electron Microscope
SILLS	Signal Integration for Laboratory Laser Systems
SIMS	Secondary Ion Mass Spectrometer
T	Temperature
TTG	Tonalite–Trondhjemite–Granodiorite
TW	Tera-Wasserburg
V-SMOW	Vienna Standard Mean Ocean Water
XRF	X-Ray Fluorescence
X-Ray μ-CT	X-Ray computed microtomography
3D	Three-dimensional

1. Introduction

1.1 Scope and structure of the thesis

The core of this PhD thesis, set up in collaboration with the Nordic Volcanological Center (NordVulk) in Reykjavik, focuses on the controversy of silicic magma generation in basalt-dominated oceanic settings. To address this problem, I targeted on the one hand the Neogene silicic volcanic systems in the remote valleys around Borgarfjörður Eystri in northeast Iceland, where I employed an integrated petrological, textural and micron-scale isotope approach to produce a quantitative analysis of rhyolite petrogenesis. On the other hand, during the course of my PhD study the submarine 2011-2012 eruption at El Hierro, Canary Islands, occurred. This eruption produced high-silica fragments during a predominantly basaltic eruption and offered the opportunity to broaden my studies to include another tectonic setting and a recent example of processes that lead to formation of high silica compositions. The El Hierro effort was funded by a scholarship I received from the European Science Foundation (ESF) through the MeMoVolc research network (Measuring and Modelling of Volcano Eruption Dynamics; www.memovolc.fr).

For this reason, this thesis is concerned with two geological regions, including three scientific articles related to northeast Iceland and two scientific articles that relate to the El Hierro 2011-2012 eruption. The first part of the thesis investigates a cluster of volcanic complexes in northeast Iceland (presented in paper I) and addresses the controversy of silicic magma genesis and related tectonic, igneous and hydrothermal processes within Iceland's upper crust (paper II and IV) that plausibly are analogous to continental crust formation on early Earth (paper III). Ultimately, this effort contributes to the long-standing petrological dilemma of how silicic continental crust was initially created on early Earth. The second part of this thesis focusses on processes of magma-crust interaction in the basaltic ocean island of El Hierro, particularly on the process of melting and degassing of crustal materials as these are entrained into magma (papers V and VI).

1.2 Background

The composition of primary mafic magma that ascends from the mantle frequently becomes modified during ascent in response to changing physicochemical conditions while traversing crustal rocks. These processes include fractional crystallisation, crustal assimilation, magma mixing, volatile mobilisation and degassing, and are usually combined in some form (Figure 1). The processes that drive large-scale silicic magmatism in basalt-dominated oceanic provinces are enormous challenges to our understanding of magmatic processes and remain the subject of lively debate ever since bimodality of volcanic products was first recognised some 160 years ago (e.g. Bunsen, 1851; Carmichael, 1964; MacDonald *et al.*, 1987; Gunnarsson *et al.*, 1998; Charreteur *et al.*, 2013). The over-abundance and -representation of mafic and silicic volcanic suites compared to rocks of intermediate compositions (bimodality), is known as the "Bunsen-Daly gap" (Bunsen, 1851; Daly, 1925). Indeed, the occurrences of bimodal (mafic-silicic) volcanic terrains extend back to early Archean continental shields that are characterised by associations of silicic TTG (Tonalite-Trondhjemite-Granodiorite) and basaltic greenstone (Kamber *et al.*, 2005; Harrison, 2009; Cawood *et al.*, 2013). Therefore, understanding the origin of bimodal volcanism and thus silicic magmas has fundamental significance for the origin of the first continental crust (Hutton, 1974). The Bunsen-Daly gap is a problem because it is difficult to reconcile with Bowen's fractional crystallisation series that is widely thought to be the principal driver of magma differentiation (Bunsen, 1851; Daly, 1925; Bowen, 1928; Wilson, 1993; Troll *et al.,* 2005; Meade *et al.*, 2014). Closed-system fractional crystallisation is unlikely to produce large volumes of silicic rocks, because the successive removal of magmatic minerals can only produce some 5 parts of residual silicic (rhyolitic) magma at the end of a full fractional crystallisation series (i.e. staring from 100 parts of basalt). Furthermore, this fractionation series should produce a successive sequence of intermediate magma compositions along the way, which is absent in bimodal volcanic suites (e.g. Tuttle and Bowen, 1958; Marsh 1989; Wilson, 1993). Crustal recycling during partial melting and assimilation of crustal melts into magma systems have therefore been considered significant processes to contribute to the generation of evolved silicic magmas (e.g. Tuttle and Bowen, 1958; Sparks and Sigurdsson, 1977; Patchett, 1980; Huppert and Sparks, 1988; Marsh *et al.*, 1991; Bindeman and Valley, 2001; Meyer *et al.,* 2009; Annen, 2011; Meade *et al.*, 2014).

1.2.1 Silicic volcanism on Iceland

Iceland is central to the discussion of bimodal volcanism because it hosts disproportionally large volumes of silicic rocks in a basaltic oceanic setting that is influenced by mid-ocean ridge processes and the underlying Iceland mantel plume (e.g. Bunsen, 1851; Carmichael, 1964; Bjarnason 2008; Bindeman *et al.*, 2012; Charreteur *et al.*, 2013). Silicic rocks constitute 10-12 % of the total rock volume exposed on Iceland and are particularly associated with central volcanoes, where rhyolites occasionally represent up to 25 % of the eruptive products (Bunsen, 1851; Walker, 1966; Gústafsson, 1992; Gunnarsson *et al.*, 1998; Jonasson, 2007). In addition, Iceland is considered the closest geodynamic analogue for the type of processes that were active during the formation of the first continents on early Earth, and therefore offers a unique opportunity to assess how silica-rich crust (continental crust) emerged from sustained basaltic volcanism on a subduction-free early Earth prior to 4 Ga (e.g. Taylor and McLennan, 1985; Maas *et al.*, 1992; Valley *et al.*, 2002; Kamber *et al.*, 2005; Martin *et al.*, 2008; Harrison, 2009; Cawood *et al.*, 2013; Reimink *et al.*, 2014).

It has been hypothesised that Icelandic rhyolites are generated in a number of ways, including (1) closed system fractional crystallisation that progressively drives magma towards evolved compositions, (2) partial melting of hydrated basaltic crust at shallow levels, or (3) a combination of assimilation and fractional crystallisation (AFC) (e.g. Carmichael, 1964; MacDonald, *et al.*, 1987; Martin *et al.*, 2011; Bindeman *et al.*, 2012; Charreteur *et al.*, 2013). Silicic rocks formed outside the active rift zones in Iceland have frequently been attributed to fractional crystallisation from a mafic parent (e.g. Hards, 2000; Prestvik *et al.*, 2001; Selbekk and Trønnes, 2007; Schattel *et al.*, 2014). One of the best-known examples is Thingmúli volcano in north-east Iceland that in the seminal descriptions by Carmichael (1964) shows a rock suite from basaltic to intermediate and silicic compositions. Carmichael interpreted this rock suite to represent a full differentiation series produced by fractional crystallisation, and this Thingmúli trend is thereafter known as the tholeiitic compositional series in Iceland (e.g. Jakobsson, 2008).

The origin of silicic rocks within the zones of active rifting has, on the other hand, commonly been proposed to involve processes of crustal assimilation (e.g. Sigurdsson and Sparks, 1981; Martin and Sigmarsson, 2010; Schattel *et al.*, 2014). Here the unique tectonic setting combined with concentrated magmatic activity on Iceland may allow for partial melting and recycling of the hydrothermally altered Icelandic crust, which can thus lead to the formation of silicic rocks (Figure 1, e.g. MacDonald *et al.*, 1987; Sigurdsson and Sparks, 1981; Marsh *et al.*, 1991; Gunnarson *et al.* 1998; Bindeman *et al.*, 2012). Conditions for voluminous partial melting of the Icelandic crust are thought to become optimised by the rapid burial of rift zone basalts during crustal accretion, subjecting basalts to extensive hydrothermal alteration and subsequent prograde burial metamorphism (Palmason, 1986). Recycling of Icelandic crust by localised partial melting is driven by the continuous propagation and relocation of the rift zone(s) into older crust that is both hot and weakened (Helgason 1985; Marsh *et al.*, 1991; Martin and Sigmarsson, 2010; Bindeman *et al.*, 2012). This crustal recycling in Iceland is supported by observations of oxygen isotopic diversity and disequilibrium textures in olivine and zircon crystals, which reflect anatexis of an isotopically heterogeneous crust (Gurenko and Chaussidon, 2002; Bindeman *et al.*, 2008; 2012). The diversity of inherited crystals has moreover been linked to contrasting oxidation conditions between different tectonic settings (rift and off-rift volcanism) that control the amount of magma-crust interaction (Schatell *et al.*, 2014; Gurenko *et al.*, 2015).

In addition to these widely recognised hypotheses, there may be an additional way of generating silicic rocks in Iceland. The abnormal crustal thicknesses, the unique geochemical rock signatures and the recovery of ancient zircon grains in some regions on Iceland, have led to the hypothesis of an old, submerged continental or oceanic sliver beneath Iceland (Schaltegger *et al.*, 2002; Foulger, 2006; Torsvik *et al.*, 2015). Partial melting and recycling of such an underlying crustal fragment could in part explain voluminous silicic magma generation in Iceland.

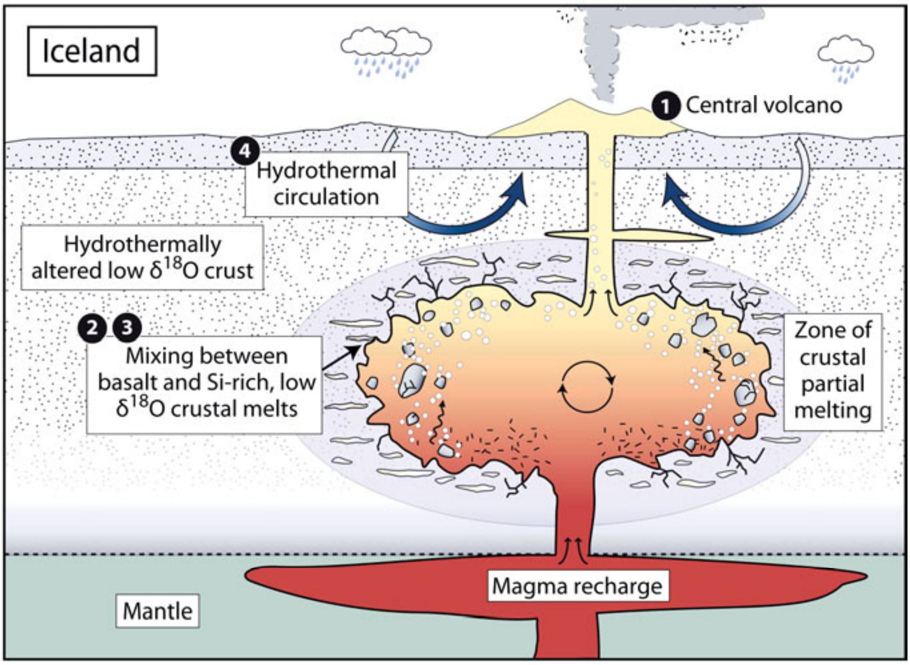

Figure 1. Schematic sketch of an Icelandic magma system that summarises the various processes that are examined in detail in this thesis (not to scale). *1)* Paper I introduces the focus of our research in Iceland and provides a geological description of our study area in northeast Iceland. *2)* Paper II focuses on crustal assimilation in Icelandic magmas as a way to produce voluminous silicic rocks. *3)* Paper III uses zircon crystals to reinforce the analogue between crustal processes occurring on early Earth and those in northeast Iceland. *4)* Paper IV investigates oxygen isotope signatures caused by infiltrating meteoric waters in hydrothermal circulation cells in Iceland (blue arrows).

1.2.2 Magma-crust interaction at El Hierro

The attribution of crustal contamination to magma diversity in volcanic systems is well documented globally. In general, crustal contamination is more frequently recognised in continental volcanic systems than on ocean islands and mid-ocean ridges, which is mainly because of the thinner and more refractory ocean crust compared to continental settings (e.g. Davies *et al.*, 1998; Hildreth and Moorbath, 1998; Davidson *et al.*, 2005). However, it has become increasingly apparent that ocean island magmas are generated with contributions from crustal components during internal island recycling (e.g. Bohrson and Reid, 1997; Gee *et al.*, 1998; Wolff *et al.*, 2000; Troll and Schminke, 2002; Hansteen and Troll, 2003; Troll *et al.*, 2012). The extent to which magma interacts with surrounding crustal rocks is controversial and typically recognised from chemical or isotopic signatures of erupted rocks or minerals. Direct evidence of crustal assimilation comes from xenoliths that are frequently hosted in igneous rocks (Clarke *et al.*, 1998; Shaw, 2009). These xenoliths are the remnants of foreign crustal fragments torn off the walls of the crustal magma reservoir and conduits, incorporated into the magma and heated above their solidus temperature. Concomitant to partial melting of the crustal fragments, the contained volatile components may exsolve into a gas phase and cause usually intense vesiculation in the upper crust (Figure 2). Continued vesicle growth and concentration during devolatilisation of the crustal fragments drives xenolith expansion and frothy (pumiceous) textures. The vesicularity itself is a direct evidence of the volume fraction of gas exsolved from the partially melting rock (Thomas *et al.*, 1994; Mangan and Cashman, 1996). Vesicle textures are fingerprints of the degassing history, and therefore offer insight into the evolutionary processes during xenolith vesiculation that help to shed light on its original source protolith (cf. Song *et al.*, 2001; Polacci *et al.*, 2008; 2009).

The 2011-2012 eruption at El Hierro ejected vesiculated crustal xenoliths that escaped complete assimilation into the magma and were found floating on the sea (Carracedo *et al.*, 2012a; Meletlidis *et al.*, 2012; Perez-Torrado *et al.*, 2012; Troll *et al.*, 2012). These xenoliths represent frozen snapshots of magma–crust interaction and offer a unique first-time chance to observe and investigate these processes in real time during active ocean island volcanism (Troll *et al.*, 2012). Frothy xenoliths such as those ejected during the El Hierro eruption have been observed elsewhere, specifically on the Canary Islands, but have been scarcely recognised until this event brought them to the attention of the scientific community. Moreover, the occurrence of silica-rich frothy El Hierro xenoliths enclosed within a basaltic carapace disclosed one way to create silica-rich partial melts in a basaltic eruption, which, on a much wider scale, may help to understand the processes that lead to the occurrence of silicic rocks in Iceland.

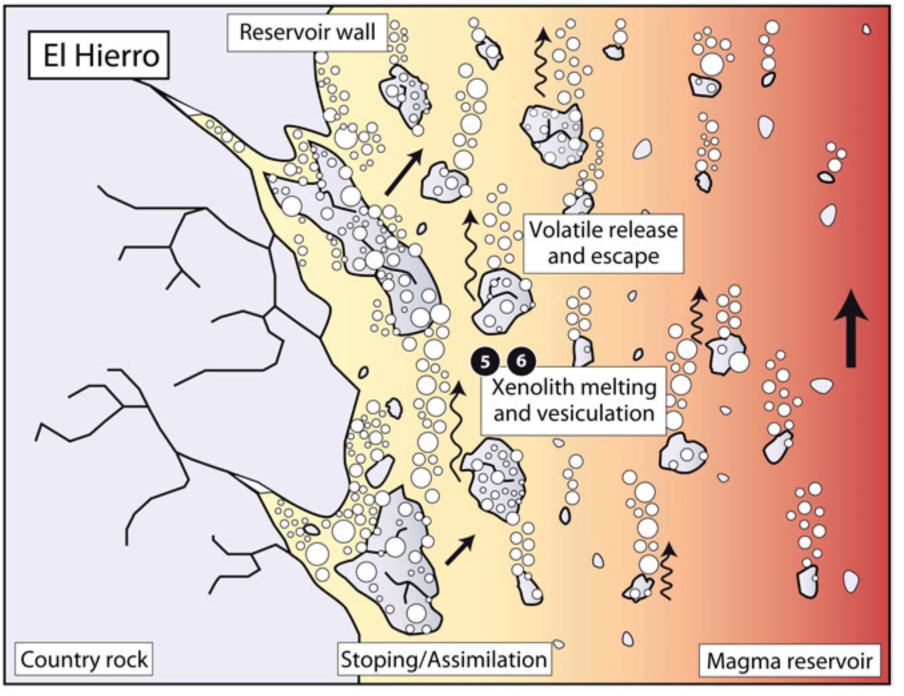

Figure 2. A diagram of the interactive processes that occur at the magma-crustal interface, with application to El Hierro. Fracturing of country rock causes crustal xenoliths to become entrained into the magma, which partially melt and degas as they become heated (i.e. forming xeno-pumice). Xeno-pumice may then erupt with the magma or disintegrate to become fully assimilated in the magma. *5)* Paper V describes the floating xeno-pumice that occurred during the early days of the El Hierro submarine eruption in 2011 and examines xeno-pumice using geo-chemical and microscopic methods (for melting and degassing phenomena) to determine their derivation. *6)* Paper VI presents a dedicated study of three dimensional vesiculation textures of the xeno-pumice using synchrotron 3D micro-CT to further investigate degassing behaviour and source material.

2. Geological setting

2.1 Iceland

Iceland is located slightly south of the Arctic Circle and is the subaerial expression of a mantle anomaly (Iceland plume) that is interacting with the spreading Mid-Atlantic Ridge. Iceland forms the most recent addition to the North Atlantic Igneous Province (NAIP) that emerged in response to the opening of the North Atlantic about 62-54 Myr ago, and which has been attributed to mantle anomaly impact under central Greenland (i.e. proto-Icelandic plume; Saunders *et al.*, 1997). The tectono-magmatic setting in Iceland is complex and characterised by two parallel rifts that are connected by transform faults, together forming the mature rift (Sæmundsson, 1979; Hardarson *et al.*, 1997; Thordarson and Larsen, 2007). The relative westward drift of the lithospheric plate boundary over the Iceland plume has forced recurring rift relocations that caused volcanic systems to shut down as magma supply dwindled and new systems became established more central to the rift (e.g. Helgason, 1985; Martin *et al.*, 2011).

The Icelandic rift system consists of several volcanic zones that in turn are composed of multiple volcanic systems, which are usually characterised by a fissure swarm and a central volcano (Sæmundsson, 1979). Present-day magmatic activity centers along three principal rift zones, the Northern Volcanic Zone (NVZ), Eastern Volcanic Zone (EVZ) and Western Volcanic Zone (WVZ), as well as the off-rift Snæfellsnes Volcanic Zone (SVZ) and rift-flank Öræfajökull Volcanic Zone (ÖVZ, Figure 3; Sigmundsson, 2006). Together these form the < 0.8 Ma Neovolcanic zone. Adjacent to both sides of the neovolcanic zone are successively older rocks, with the oldest dated rock occurring in the extreme northwest Iceland (17-16 Ma; e.g. Moorbath *et al.,* 1968; Riishuus *et al.*, 2013).

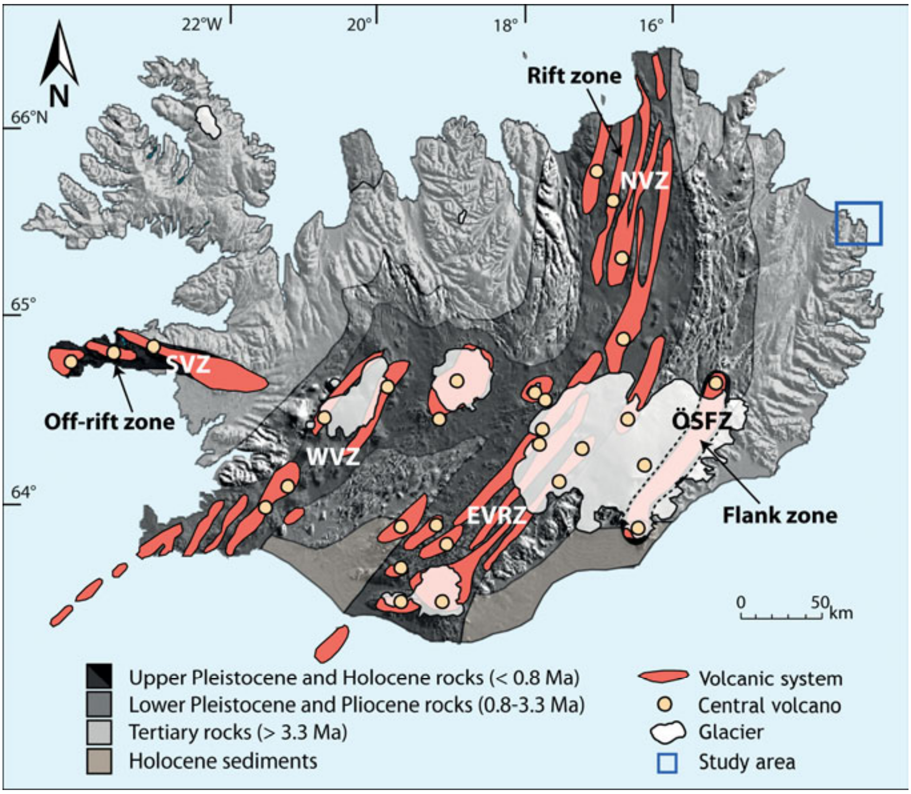

Figure 3. Geological map of Iceland showing the volcanic zones with their respective volcanic systems that together compose the rift system of Iceland. The northern, western and eastern volcanic zones (NVZ, WVZ and EVZ) form the mature rift, while the Snæfellsnes and Öræfajökull volcanic zones (SNVZ and ÖVZ) are flank zones that differentiate on their geochemical composition. The Icelandic crust is youngest in the rift zones and maturates towards East and West, with the oldest rocks (17-16 Ma) found in the far northwestern fjords. The study area around Borgarfjörður Eystri in northeast Iceland (13-12 Ma) is boxed.

The region of glacially eroded fjords between Borgarfjörður Eystri and Loðmundarfjörður in northeast Iceland is the research area for the bulk of this thesis and comprises a spectacular cluster of fossil Neogene central volcanoes (13-12 Ma), including Dyrfjöll, Breiðavik, Kækjuskörð, and Herfell, that lie embedded within a tick lava pile of regional flood basalts (e.g. Walker, 1963; Gústafsson, 1992; Óskarsson and Riishuus, 2013). The contemporaneously active central volcanoes produced rhyolite lava flows, subvolcanic intrusions and pyroclastic density currents (ignimbrite deposits) to an amount unusual for Iceland (Gústafsson et al., 1989; Burchardt et al., 2011; Martin et al., 2011; Óskarsson and Riishuus, 2013). The large volumes and geographically extensive ignimbrite sheets, as well as several collapse calderas in the region, bear witness to the explosive past of the Neogene volcanism in this region, with the famed Hvítserkur ignimbrite being one of the largest ignimbrite sheets in the country and a photogenic landmark of north-east Iceland (see Figure 4). The volcanism around Borgarfjörður Eystri, produced Neogene silicic rocks in excess of 20 vol. %, exceeding the usual ≤ 12 vol. % on Iceland, and is characterised by a strongly bimodal (mafic–felsic) distribution (cf. Gústafsson et al., 1989; Gústafsson, 1992). This area represents a complete cycle of silicic volcanism that is constrained from beginning to end by the enveloping groups of fissure-fed flood basalts. The exposed silicic central volcanoes therefore provide an extraordinary opportunity to constrain the processes and timescales of large-volume silicic magmatism in an otherwise basaltic province.

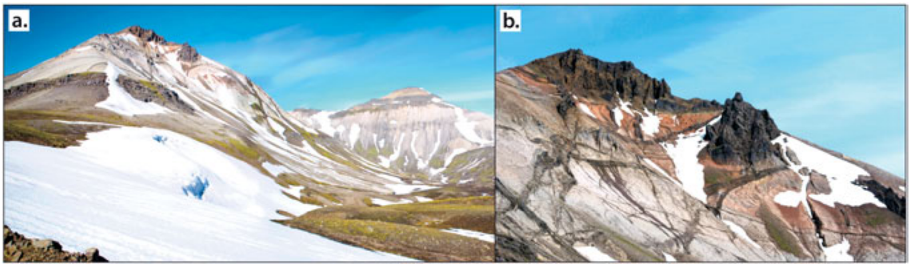

Figure 4. a) The mountain Hvítserkur in the Breiðavik volcano consists of one large caldera-filling ignimbrite sheet that is crosscut by a system of basaltic dykes. b) Close-up of the summit region. The beige to pink ignimbrite is overlain by basaltic hyaloclastites and pillow breccias.

2.2 El Hierro, Canary Islands

The island of El Hierro in the Canary archipelago is the westernmost, smallest and also the youngest part of the East-West aligned chain of seven volcanic islands (Figure 5; Guillou *et al.*, 1996). The Canary Islands get younger towards the west, from ca. 22 Ma on Fuerteventura to ca. 1.2 Ma on El Hierro (Carracedo *et al.*, 1998), which is widely attributed to an underlying mantle plume (e.g. Carracedo *et al.*, 2001; Geldmacher *et al.*, 2005; Zaczek *et al.*, 2015). El Hierro is characterised by a trihedral shape of three convergent ridges of volcanic cones that are separated by wide embayments, which are the scars of several massive gravitational landslides that truncate the flanks of the island (Carracedo, 1996; Carracedo *et al.*, 2001; Carracedo *et al.*, 2015; Carracedo and Troll, 2016). The most recent giant landslide in the Canary archipelago is thought to be the collapse of El Hierro's northern flank, leaving behind the spectacular El Golfo embayment with a 1400 m-high and nearly vertical escarpment (e.g. Carracedo *et al.*, 2001; Manconi *et al.*, 2009; Longpré *et al.*, 2011; Carracedo and Troll, 2016). The volcanic history of the island largely comprises three main episodes of growth that are parted by gravitational collapse events (Guillou *et al.*, 1996; Carracedo *et al.*, 2001). These main volcano-stratigraphic units are i) the Tinor volcano that formed the first subaerial rocks, ii) the El Golfo volcano which contains volumetrically subordinate trachybasalt to trachyte flows and block-and-ash deposits at the terminal stages of the volcano, and iii) recent rift volcanic activity with basaltic, picritic and ankaramitic products. The geology of El Hierro is characteristic of an ocean island that is still in its early stage of shield construction, which has taken place during the past one million years approximately. El Hierro is the subaerial edifice of the volcanic shield that continues below sea level as a submarine ridge down to about 3700 m depth. The seafloor below is composed of ≤ 1 kilometer of sedimentary rocks overlying Jurassic oceanic crust (Ranero *et al.*, 1995; Gee *et al.*, 2001; Montesinos *et al.*, 2006; Zaczek *et al.*, 2015; Carracedo and Troll, 2016).

No prior historic volcanic activity on El Hierro was known when a submarine eruption commenced on October 10, 2011 off the island’s southern coast. The eruption was preceded by several months of seismic unrest that was associated with vertical and lateral magma movement, and the eruption continued for several months, but remained submarine for the whole period until activity finally ceased in March 2012 (e.g. Carracedo *et al.*, 2012ab). Surface expressions of this eruption included a floating plume of stained water, together with strong bubbling and degassing, and floating lava bombs. Two distinct types of lava bombs were encountered at separate stages of the eruption. 1) During the first week of the eruption, abundant decimetre sized, light-coloured pumiceous, high-silica bombs, enclosed by a basanite carapace were found floating on the sea (“xeno-pumice”), whereas 2) entirely basanitic “lava balloons”, usually with hollow interiors and exceeding 1 metre in size, occurred throughout the eruption and have been observed during submarine eruptions elsewhere (Figure 5; cf. Siebe *et al.*, 1995; Gaspar *et al.*, 2003; Kueppers *et al.*, 2012). The remarkable pumiceous silicic xenoliths are known from other eruptions in the Canaries (e.g. Schmincke and Graf, 2000; Hansteen and Troll, 2003; Aparicio *et al.* 2010), however, the origin of the El Hierro xeno-pumice was the source of considerable debate at the time, with implications for the hazard assessment of the ongoing eruption (see Meletlidis *et al.*, 2012; Perez-Torrado *et al.*, 2012; Troll *et al.*, 2012; 2015; Carracedo *et al.*, 2015; Del Moro *et al.*, 2015; Rodriguez-Losada *et al.*, 2015).

The 2011-2012 El Hierro eruption was the first time a submarine eruption in the Canary Islands was monitored from its early precursors to termination (e.g. López *et al.*, 2012; Pérez-Torrado *et al.*, 2012; Gonzales *et al.*, 2013; Longpré *et al.*, 2014). The eruption provided not only a valuable opportunity to advance our understanding of the accretion processes that form the Canary Islands and of the structure of the underlying crust, but from a societal perspective it also brought about enormous advances for improving volcanic crisis management (Carracedo *et al.*, 2015).

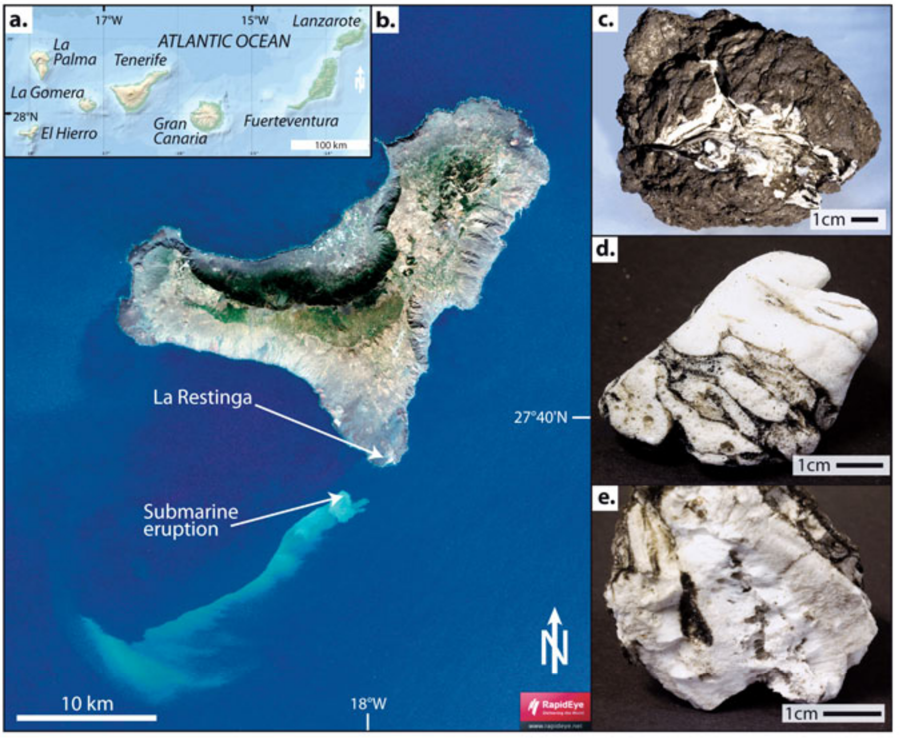

Figure 5. a) Topographic map of the Canary Islands off the coast of northwestern Africa. El Hierro is the youngest and westernmost island in the archipelago. b) True colour RapidEye satellite image of El Hierro island, captured by the Moderate Resolution Imaging Spectroradiometer (MODIS) aboard the Terra satellite on December 21, 2011. The stain on the ocean caused by the submarine eruption can be seen SSE of El Hierro. Image credit: Jeff Schmaltz MODIS Land Rapid Response Team, NASA GSFC. c) - e) A selection of xeno-pumice hand specimen that erupted during the early days of the eruptive event. The xeno-pumice bombs exhibit intense mingling and mixing between the enclosing basanite carapace and the light coloured pumiceous cores, see c) and d). The xeno-pumice samples appear fritted and are highly porous with particularly vesiculated core regions (see e), which allowed them to float on the sea surface.

3. Methodology

This thesis utilises a variety of techniques, including major and trace element analyses of whole-rock samples, oxygen isotope determination of whole-rocks and minerals, zircon cathodoluminescence, U-Pb geochronology, trace and rare earth element (REE) analyses in zircons, as well as rock volume calculations from geological maps, and three-dimensional (3D) imaging using synchrotron tomography techniques. A summary of the analytical techniques is provided below.

3.1 Sample preparation

Rock samples were first of all washed and cut to remove weathered surfaces, whereupon they were crushed to coarse rock fragments in a jaw crusher and milled to whole-rock powders in an agate ball mill at NordVulk. Feldspar and pyroxene minerals were handpicked from selected rock samples at Uppsala University (UU). Between 2 to 5 kg of crushed rock from 15 samples, as well as collected samples of fluvial river sands were used to extract zircon crystals. The zircon fractions were separated by Geotrack International Pty Ltd, Australia. Following a conventional separation procedure of crushing, milling and sieving, the ≤ 250 µm grain fractions was passed over a Wilfley table. Concentrated zircon separates were produced using magnetic separation and heavy liquids. Final purification was performed by hand under a stereomicroscope at Uppsala University.

3.2 Whole-rock major, trace and rare earth elements

Major and trace elements of samples presented in papers II, III and IV were analysed from powdered pristine rock chips at ACME Labs Ltd, Vancouver, Canada. Major oxides were determined from rock powders by X-Ray Fluorescence (XRF), while trace and rare earth elements were measured by inductively coupled plasma-mass spectrometry (ICP-MS) after preparation by multi-acid digestion. Loss on ignition (LOI, i.e. volatile content) was measured by the weight difference after ignition of sample splits at 1000 °C. Measurements of internal standards and duplicates monitored analytical reproducibility.

3.3 Oxygen isotopes in whole-rocks and minerals

Oxygen isotope in whole-rocks and mineral separates are presented in papers II and IV and were measured at the University of Cape Town (UCT), South Africa, using both conventional and laser fluorination methods and following the extraction procedure outlined in Vennemann and Smith (1990) and Fagereng *et al.* (2008). All values are reported in the standard δ-notation, where $\delta = (R_{sample}/R_{standard} - 1) \times 1000$, and $R = {}^{18}O/{}^{16}O$.

Isotope ratios were measured off-line using a Thermo DeltaXP mass spectrometer in dual inlet mode, and a conventional silicate extraction line with externally heated Ni vessels, which are described in detail in Harris and Ashwal (2002) and Curtis *et al.* (2013). For the conventional oxygen isotope analyses ~10 mg powdered sample was oven-dried at 50 °C and degassed under vacuum on a conventional silicate line at 200 °C. Silicates were reacted with ClF_3 (Borthwick and Harmon, 1982) and the liberated O_2 was converted to CO_2 using a hot platinised carbon rod (Vennemann and Smith 1990; Harris and Ashwal 2002; Fagereng *et al.,* 2008). Samples were run on the vacuum line along with duplicate samples of the internal quartz standard MQ ($\delta^{18}O$ = 10.1 ‰) that was used to calibrate the raw data to the SMOW scale (Standard Mean Ocean Water; e.g. Sharp, 2007). During the course of this study, the analytical error was 0.16 ‰ (2σ) for all samples.

The O-isotope ratios of mineral separates (n = 8) in paper IV were analysed using the laser fluorination vacuum line at UTC and measured on purified O_2 gas. Full analytical details are described in Harris and Vogeli (2010). The internal standard MON GT ($\delta^{18}O$ = 5.38 ‰, Harris and Vogeli, 2010) was used to calibrate the raw data and correct for drift in the reference gas. The long-term average difference in $\delta^{18}O$ values of duplicates of MON GT analysed during this study was 0.13 ‰, and corresponds to a 2σ error value of 0.16 ‰. MON GT was recalibrated against the UWG-2 garnet standard of Valley *et al.*, (1995), using the revised $\delta^{18}O$ value of 5.38 ‰ and assuming a $\delta^{18}O$ values of 5.80 ‰ for UWG-2.

3.4 Zircon cathodoluminescence

The suite of zircons presented in paper II was morphologically and texturally characterised by cathodoluminescence (CL) imaging using a Hitachi scanning electron microscope (SEM) at the NordSIM ion microprobe facility, Swedish Museum of Natural History in Stockholm. The detrital zircon sample suite used in paper III was CL-imaged using the field emission source JEOL JXA-8530F Hyperprobe at CEMPEG (Centre for Experimental Mineralogy, Petrology and Geochemistry), Uppsala University, Sweden.

3.5 Zircon U-Pb geochronology

The single-grain, high precision U-Pb radiometric analyses of zircons presented in paper II were obtained using the Cameca IMS1280 ion microprobe (secondary ion mass spectrometer, SIMS) at NordSIM (Figure 6). Detailed analytical methods are described in Whitehouse *et al.* (1999) and Whitehouse and Kamber (2005). Fracture and inclusion free cores and internal zones of individual crystals were targeted for spot analyses using a 15 μm diameter ion beam. The analyses used 12 cycles and the data were calibrated by employing repeated analyses of the zircon 91500 standard (Wiedenbeck *et al.*, 1995). For each unknown, ^{207}Pb-corrected ages were calculated from common-Pb uncorrected ratios using *ISOPLOT 4.15* (Ludwig, 2003), with decay constants following the recommendations of Steiger and Jäger (1977). The ^{207}Pb-corrected ages were used in all data interpretations, assuming a common Pb composition that projects through the uncorrected data point onto Tera-Wasserburg concordia. Uncertainties are presented at the 2σ confidence level (disregarding decay constant errors). All analyses were carefully screened and those that were not deemed robust, either because of high standard deviation errors or unsuccessful spot locations (e.g. straddling grain boundaries, cracks or inclusions), were not considered further during data processing. Around 90 % of the analyses plot within a 5 % confidence limit of the concordia, and ca. 10 % outliers were excluded from further age calculations. Rock unit ages were derived from the weighted average of the individual ^{207}Pb-corrected zircon ages. The MSWD (mean square weighted deviation) value of each average is an indication of how likely it is that the assigned analytical errors explain the scatter. For MSWD < ~1.5 the age is robust, however, the overall error allows for larger MSWD values and so can be considered realistic as well, although producing larger errors as a result.

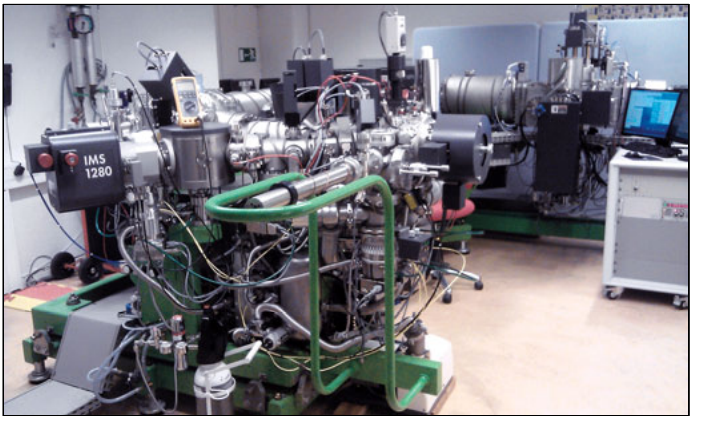

Figure 6. The Cameca IMS1280 ion probe at the NordSIM facility within the Department of Geosciences, Swedish Museum of Natural History, Stockholm.

3.6 Zircon oxygen isotope analyses

The zircons subjected to U-Pb geochronology dating in paper II were also analysed for oxygen isotope composition. Each oxygen spot location was set within the dated growth zones and as close as possible to the U-Pb SIMS pits. In the case of large zircon crystals that showed prominent core-rim zoning, $\delta^{18}O$ crystal transects were also carried out. SIMS oxygen isotope analyses follow the procedure outlined in Nemchin *et al.* (2006) and Whitehouse and Nemchin (2009). A 20 keV Cs^+ primary beam of ca. 2.5 nA was used in critically-focused mode together with a 5 µm raster to sputter a ~10 µm wide sample area. A normal incidence low energy electron gun provided charge compensation. The runs comprised a pre-sputter period of 90 seconds with a raster of 20 µm, and field aperture centering using ^{16}O signal, which was followed by 64 seconds of data acquisition using two Faraday detectors in the multi-collector system that operates at a common mass resolution of ca. 2500. The secondary magnet field was regulated at high precision using a Metrolab NMR teslameter. Instrumental mass fractionation was determined using the zircon 91500 standard (Wiedenbeck, 2004), and the ratios of the unknowns were corrected accordingly. Two analyses of the reference material were run between every six unknown analyses. Temora 2 zircon was used as secondary reference standard (Black *et al.,* 2004) and one analysis was run between every 12 analyses (i.e. after every second set of zircon 91500 analyses). Internal precision (1σ mean) on $\delta^{18}O$ for both the 91500 reference material and the unknowns had an average value of ± 0.24 ‰ over the 4 analytical sessions. The external error on zircon 91500 ranged from ± 0.15 ‰ to 0.25 ‰ (RSD; n = 125), and external errors were propagated into the overall analytical error for each analysis.

3.7 Zircon trace and rare earth elements (LA-ICP-MS)

Paper III focuses on trace element concentrations in zircons that were determined by laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) using a 193 nm Resonetics ArF excimer laser coupled to a Thermo Element XR ICPMS at the Institute of Geochemistry and Petrology, ETH Zurich, Switzerland. The laser spot size was 30 µm and its output energy was maintained at around 3.5 J/cm^2. To monitor instrumental drift the internal standard NIST 612 was used, and GSD-1G was employed as a secondary standard. Raw data were reduced using the MATLAB-based program SILLS (Guillong *et al.*, 2008). Only stable signals were used during integration of the data. Aberrations in signal count are caused by analyses hitting inclusions with different elemental concentrations e.g. apatite (see Figure 7), and these were consequently omitted. Analytical precision was quantified from long term (> 3 year) assessment of homogeneous materials, which shows that analytical uncertainty is << 5 % relative to the stated abundance for elemental concentrations significantly above the limits of detection (i.e. 2x LOD).

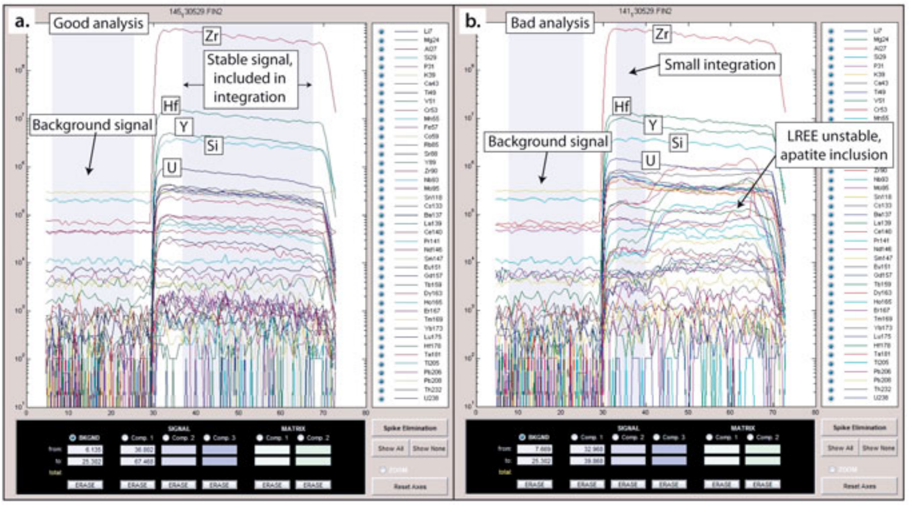

Figure 7. Snapshots of the zircon trace element data produced by LA-ICP MS that are reduced using the SILLS program. a) A good analysis is characterised by a broad and stable signal, whereas b) analyses hitting inclusions generates unstable signals with suddenly increasing elemental signals, yielding a restricted integration range and the analysis usually becomes excluded from the dataset.

3.8 3D structural modelling reconstructing pre-erosional rock volumes

Silicic rock volumes in the greater Borgarfjörður Eystri area that are presented in paper II were calculated with the *ArcGIS* software, using a digital elevation model (cell size 20x20 meters) and a geological map of the area (Gústafsson *et al.*, 1989; Gústafsson, 1992), which was revised based on our fieldwork and mapping in the region. This approach provided post-erosional volume estimates. Pre-erosional volume estimates were assessed using two different approaches in the software *Move*^{TM} by Midland Valley Ltd. The first method reconstructed the top and bottom surfaces of a prominent ignimbrite unit in the area, by approximating the palaeo-land surface with a horizontal surface at 900 m above sea level (m.a.s.l.; Gústafsson, 1992). However, this calculation disregarded Neogene topography and the irregular distribution of silicic rocks, which are often located at higher elevations than the assumed 900 m.a.s.l.

A more accurate estimate of the pre-erosional volume of silicic rocks was derived from present-day exposure of the Hvítserkur ignimbrite that has well-constrained bottom and top limits. Its original volume was constrained by extrapolation to fill the whole caldera where it originally was deposited. Neglecting the amount of erupted material deposited outside the caldera, the minimal reconstructed volume is 7.5 times greater that today's exposure. Assuming that the Hvítserkur ignimbrite is representative of the whole study area, these volume estimates were extrapolated to derive pre-erosional rock volumes in the region. Further details of these volume calculations are available in paper II.

3.9 Synchrotron X-ray microtomography

Paper VI in this thesis utilises internal three-dimensional imaging of xeno-pumice samples that were performed by X-ray computed microtomography (μ-CT) at the SYRMEP beamline (Tromba *et al.*, 2010) and at the TomoLab station (Zandomeneghi *et al.*, 2010) of the Elettra synchrotron light laboratory in Trieste (Italy, Figure 8a). Small xeno-pumice cubes (1 x 1 x 1 cm) were scanned using a monochromatic, nearly-parallel X-ray beam in phase-contrast mode at a distance of about 23 m from the source (Figure 8b). The μ-CT experiments collected about 1440 radiographs per sample over a scan angle of 180°, using a sample-to-detector distance of 200 mm and beam energy of 25 to 38 KeV. The detector was a 12 bit water-cooled CCD camera (Charge Coupled Device), with a pixel size of 9 x 9 μm and 12 x 18 mm field of view. The custom-developed software *Syrmep_tomo_project 4.0* (Montanari, 2003) was used to reconstruct the collected radiographs into 2D axial slices, which is based on the filtered back-projection algorithm of Kak and Slaney (1987).

A subset of samples was analysed in the TomoLab station of Elettra using a microfocus phase-contrast X-ray μ-CT, which provides a polychromatic beam of cone shaped geometry. Using a Voltage of 80 - 100 kV, 2400 radiographs were collected over a scan angle of 360°. A water-cooled, 12 bit CCD camera was used as detector, with an effective pixel size of 25 x 25 μm and a 50 mm x 33 mm maximum field of view. The pixel size (resolution) of the CT scans ranges from 5 to 7.1 μm, depending on the set source-to-sample (9 - 10 cm) and source-to-detector distance (35 - 45 cm). Further methodological details are provided in Ketcham and Carlson (2001) and Polacci *et al.* (2010). The slice reconstruction was performed by the commercial software *COBRA* (Exxim, USA).

Digital volumes of all samples were rendered and processed in the freeware *ImageJ* (Abramoff *et al.*, 2004) and the commercial software *VGStudio MAX 2.0*®. Segmentation algorithms, as well as the morphological and textural analyses of the samples were mainly performed with custom-developed *Pore3D* library at Elettra (Brun *et al.*, 2010), but also using the freeware plugin *BoneJ* (Doube *et al.*, 2010).

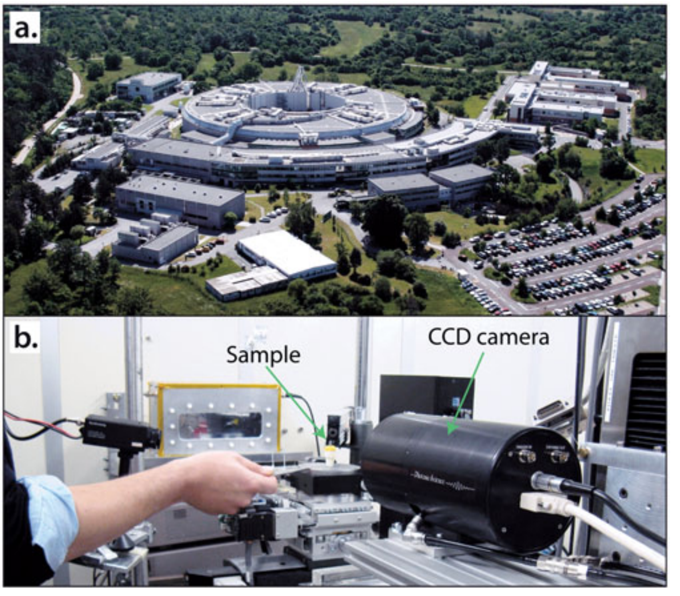

Figure 8. a) Elettra synchrotron light laboratory in Trieste, Italy (image courtesy of Elettra-Sincrotrone Trieste S.C.p.A.). b) The µ-CT experimental setup at the SYRMEP beamline of Elettra.

3.10 Other methodologies

X-ray Diffraction (XRD)

The xeno-pumice mineral assemblage presented in paper V was determined from powdered material of pristine xeno-pumice chips using a Siemens/Bruker D5000 diffractometer at the Geological Survey of Sweden (SGU) in Uppsala. More details are available in paper V.

4. Summary of the papers

4.1 Paper I

Iceland's best kept secret

The greater Borgarfjörður Eystri area in northeast Iceland hosts mountains of voluminous silicic rocks concentrated within a remarkably small area, including unusually large volumes of ignimbrite sheets that document extremely violent eruptions during the Neogene. This remote region is characterised by a pronounced compositional bimodality, and represents one of the most voluminous occurrences of silicic rocks in the whole of Iceland, but is nonetheless sparingly investigated. This area may provide an answer to the long-standing petrological dilemma of how silicic continental crust is initially created in basalt dominated settings. In this popular science article (paper I) we set the scene for an in-depth investigation that focuses on the controversy of rhyolite petrogenesis on Iceland. We present a background study of current theories of silicic magma genesis, and document our adventurous geological journey and field strategy in the remote valleys of northeast Iceland.

4.2 Paper II

Rapid generation of high-silica magmas in basalt-dominated rift settings

The processes that drive large-scale silicic magmatism in basalt-dominated provinces have been widely debated for well over a century, with Iceland taking a central place in this discussion (e.g. Bunsen, 1851; Carmichael, 1964; MacDonald *et al.*, 1987; Gunnarson *et al.*, 1998; Martin *et al.*, 2011; Charreteur *et al.*, 2013). Several hypotheses have been put forward to explain the generation of Iceland's silicic rocks, but as yet no consensus exists (e.g. Carmichael, 1964; MacDonald *et al.*, 1987; Bindeman *et al.*, 2012; Charreteur *et al.*, 2013). Understanding the causes for bimodal volcanism is not only relevant for modern rift settings, but also for advancing our general concepts of how silicic continental crust formed from the basalt-dominated magmatic environment on early Earth (e.g. Reimink *et al.*, 2014). In order to understand the processes and timescales of silicic crust generation in basaltic provinces, we investigated the silicic volcanic complexes around Borgarfjörður Eystri in northeast Iceland. This region represents the largest (> 450 km^3) complete pulse of Neogene rift-related silicic magmatism preserved on Iceland. In paper II, we use zircon geochronology and oxygen isotope geochemistry to show that the silicic magmas were generated in ≤ 2 Myr during heightened activity of the Iceland plume (Figure 9). Moreover, zircon $\delta^{18}O$ values fall dominantly below mantle values, recording up to 30 % input of low-$\delta^{18}O$ hydrothermally-altered crust at 12.5 Ma, the height of explosive activity. This event was followed by a rapid termination of silicic magma production within only 0.6 Ma, as crustal fertility declined (cf. Meade *et al.*, 2014). Since Iceland is widely considered a modern-day analogue for continental crust formation on early Earth (e.g. Reimink *et al.*, 2014), the results presented in paper II may now offer a mechanism and time-scale to help understand rapid, voluminous silicic magma generation in basalt-dominated rift settings.

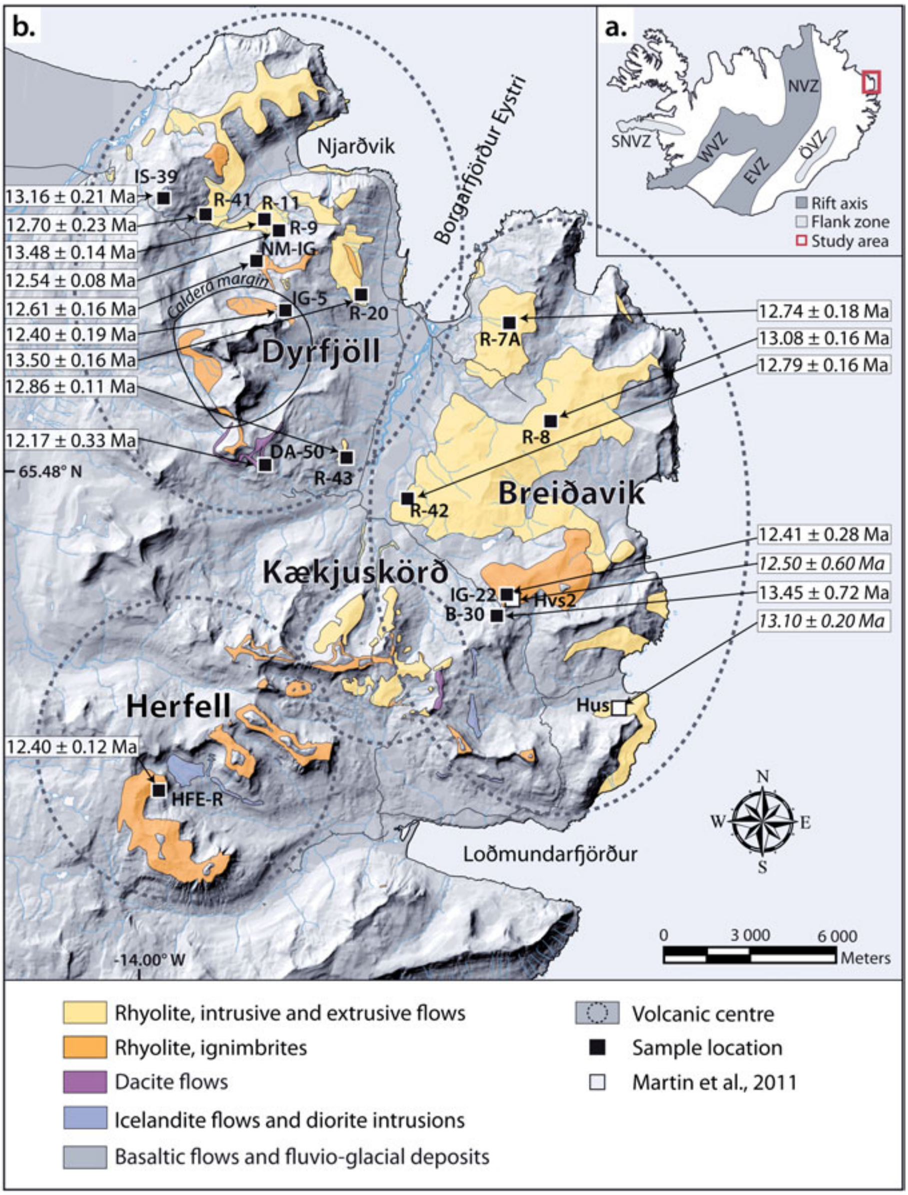

Figure 9. Silicic volcanism in Borgarfjörður Eystri region. a) Map of Iceland showing the northern (NVZ), western (WVZ), and eastern (EVZ), as well as Snæfellsnes (SNVZ) and Öræfajökull (ÖVZ) volcanic zones. Study area in northeast Iceland is boxed. b) Geological map of the Borgarfjörður Eystri region showing the amount of silicic rocks, and with dated rock samples marked. Ages for key units from the volcanic centres constrain the silicic cycle to ≤ 2 Myr duration, which includes an episode of explosive volcanism that lasted ~0.5 Myr. Literature samples are presented in light grey.

4.3 Paper III

Zircons from northeast Iceland analogous to those from early Earth

Paper III addresses the proposed analogue between northeast Iceland and processes occurring on early Earth, which was brought to attention in paper II.

Earth's earliest silicic continental crust is alleged to have formed in a dominantly basaltic, plume-dominated, subduction–free tectonic setting, while the exact processes involved remain unclear (Kamber *et al.*, 2005; Hawkesworth and Kemp, 2006; Harrison, 2009; Bédard *et al.*, 2013; Cawood *et al.*, 2013). This knowledge gap is primarily due to the near absence of a preserved rock record older than 4.0 Ga, whilst direct evidence of Hadean Earth comes principally from xenocrystic zircons that are found in only a few localities worldwide (e.g. Condie, 2007; Harrison, 2009). Defining present-day analogues therefore becomes fundamental to advance our understanding of these ancient processes. Iceland shows strikingly similar geodynamic characteristics to early Earth and has long been considered as a prime candidate for a setting that may reflect the type of processes that caused nucleation of the earliest continental crust on early Earth (e.g. Taylor and McLennan, 1985; Maas *et al.*, 1992; Valley *et al.*, 2002; Martin *et al.*, 2008; Reimink *et al.*, 2014). However, this concept of Iceland as an analogue for early Earth is disputed and was recently negated because of the significant differences between zircon trace element concentrations, Ti-thermometry and oxygen isotope values in a wide range of Icelandic samples relative to those recorded in known Hadean zircons (Carley *et al.*, 2014). The compositional diversity among Icelandic zircons that is potentially related to contrasting types of silicic magma generation in rift and off-rift settings in Iceland (cf. Bindeman *et al.*, 2012; Schattel *et al.* 2014; Gurenko *et al.*, 2015) however, suggests that analogues to early Earth are better looked for on a local scale. In paper III, we present compositional data for zircon from a region in northeast Iceland (Borgarfjörður Eystri) that hosts abnormally high proportions of silicic crust compared to the Icelandic average and formed within a backdrop of sustained basaltic volcanism. The aim of our study is to test if this region in northeast Iceland could serve as a potential analogue for early Earth processes, although there is no apparent connection between Hadean and bulk Icelandic zircons (Carley *et al.*, 2014).

We show that trace and rare earth element concentrations in zircons from Borgarfjörður Eystri frequently overlap with Hadean zircons, particularly with respect to Ti-thermometry temperatures of the zircon source (Figure 10). This leads us to conclude that, while Iceland as a whole may not be an ideal analogue for the Hadean (cf. Carley *et al.*, 2014), the silicic province around Borgarfjörður Eystri may provide a better parallel. The complex plume-related rift-flank setting that gave rise to the Neogene rocks in northeast Iceland may therefore constitute a viable geodynamic and geochemical analogue for the formation of Earth's earliest continental crust.

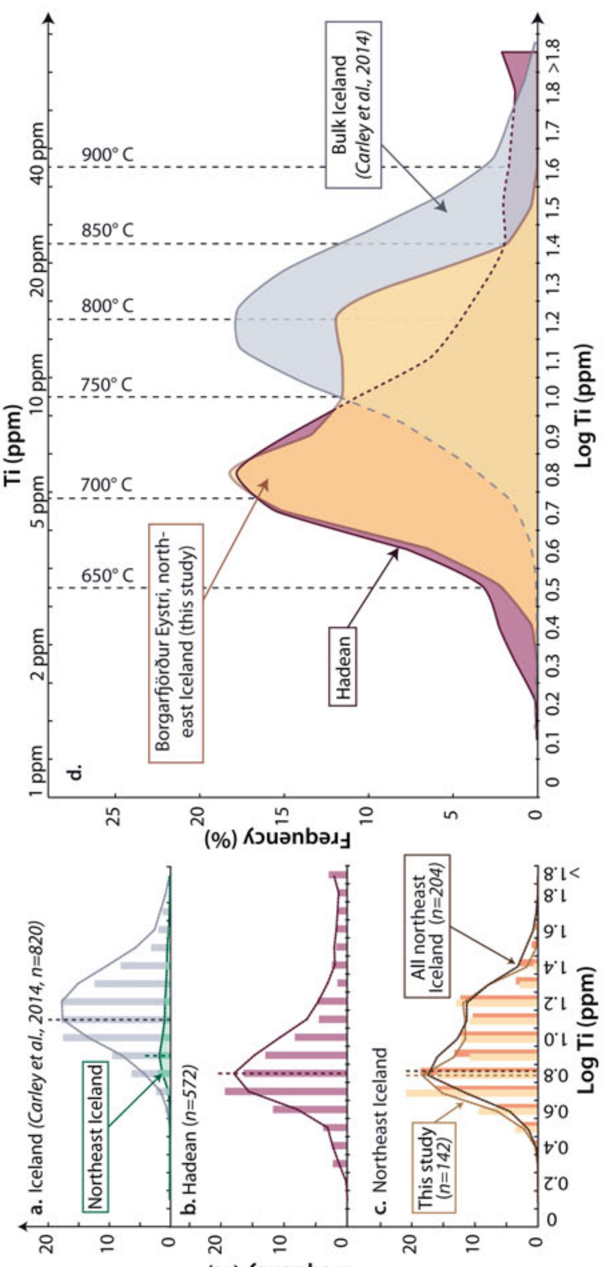

Figure 10. Histograms of log Ti content (ppm) in zircons from a) Iceland as a whole, including the extracted data from northeast Iceland in green (Carley *et al*., 2014), b) the Hadean and c) from Borgarfjörður Eystri (this study, yellow), as well as the compiled dataset from northeast Iceland (i.e. this study plus data extracted from Carley *et al*., 2014). For each dataset, a trend line was calculated from a moving average (sample window=2 data points). d) Simplified frequency curves from a)-c) are integrated with minimal magma temperatures that are estimated after Ferry and Watson (2007). The data from Borgarfjörður Eystri (this study) show a right-skewed distribution that partly overlaps with the Hadean zircons, but partly also with the regional Iceland zircon data. Notably, Borgarfjörður Eystri zircons share many characteristics with Hadean zircon data and differ from the "bulk Iceland" group, indicating a similar set of formation processes might be reflected in Hadean and Borgarfjörður Eystri zircons.

4.4 Paper IV

Origin of high whole-rock $\delta^{18}O$ values in rhyolites from northeast Iceland

The Icelandic crust is characterised by low $\delta^{18}O$ values that originate from pervasive high-temperature hydrothermal alteration by circulating ^{18}O-depleted meteoric waters (Muehlenbachs *et al.*, 1974; Hattori and Muehlenbachs, 1982; Eiler, 2001; Bindeman, 2008; Bindeman *et al.*, 2012). These deep-seated meteoric-hydrothermal systems are largely a result of Iceland's subaerial exposure at high latitudes, proximity to seawater, high rates of precipitation, in combination with high rates of volcanism and crustal extension (Eiler, 2001). In general, igneous rocks with $\delta^{18}O$ values higher than normally can be produced by closed-system fractionation from a mantle-derived magma (i.e. +5.7 to ~7 ‰; Valley *et al.*, 2005; Bindeman, 2008) have typically assimilated material altered at low temperature or undergone isotope exchange at low temperatures themselves (e.g. Bindeman, 2008; Donoghue *et al.*, 2008; 2010). Igneous rocks on Iceland that have $\delta^{18}O$ values significantly higher than unaltered oceanic crust (~5.7 ‰) are scarce (e.g. Muehlenbachs *et al.*, 1974; Hattori and Muehlenbachs, 1982; Condomines *et al.*, 1983; Macdonald *et al.*, 1987; Hemond *et al.*, 1993; Gunnarsson *et al.*, 1998; Prestvik *et al.*, 2001; Macpherson *et al.*, 2005). This paucity is mainly a result of low temperature alteration processes on Iceland that are less likely to produce high $\delta^{18}O$ values form the ^{18}O-depleted meteoric waters available, ranging from -7.7 to -15 ‰ (e.g. Árnason, 1976; Hattori and Muehlenbachs, 1982; Rozanski *et al.*, 1993). Moreover, the rate of chemical weathering on Iceland is in any case likely to be low because of the generally low ambient temperatures.

In paper IV, we report whole-rock and mineral oxygen isotope data in Neogene ignimbrites of the central volcano cluster of Borgarfjörður Eystri, northeast Iceland, that display $\delta^{18}O$ values that exceed the mantle range (Figure 11). In particular, we record $\delta^{18}O$ values that range from +15.5 to +18.5 ‰ and thus represent the highest $\delta^{18}O$ values so far recorded for Icelandic rocks. Co-existing pyroxenes in the ignimbrite samples have $\delta^{18}O$ values of +4.0 to +4.2 ‰, which are consistent with $\delta^{18}O$ values recorded in zircon from the same rock units, and confirm O-isotope equilibrium with the parent magmas at the temperature of crystallisation, while being out of equilibrium with the bulk rock $\delta^{18}O$ value. The high whole-rock $\delta^{18}O$ values of these ignimbrite samples most likely result from pervasive exchange during subsolidus hydrothermal alteration by ^{18}O-enriched water. Such alteration may have conceivably occurred *1)* in a near surface hot-spring environment at a distal end of a hydrothermal system, fed by waters that already exchanged significantly with the country rock, or *2)* between ignimbrites and

^{18}O-enriched alteration fluids produced during boiling and evaporation of standing water in former caldera lakes. Irrespective of the exact process of alteration, we confirm that a previously unrecognised, and probably highly localised, high-$\delta^{18}O$ crustal end-member exists on Iceland.

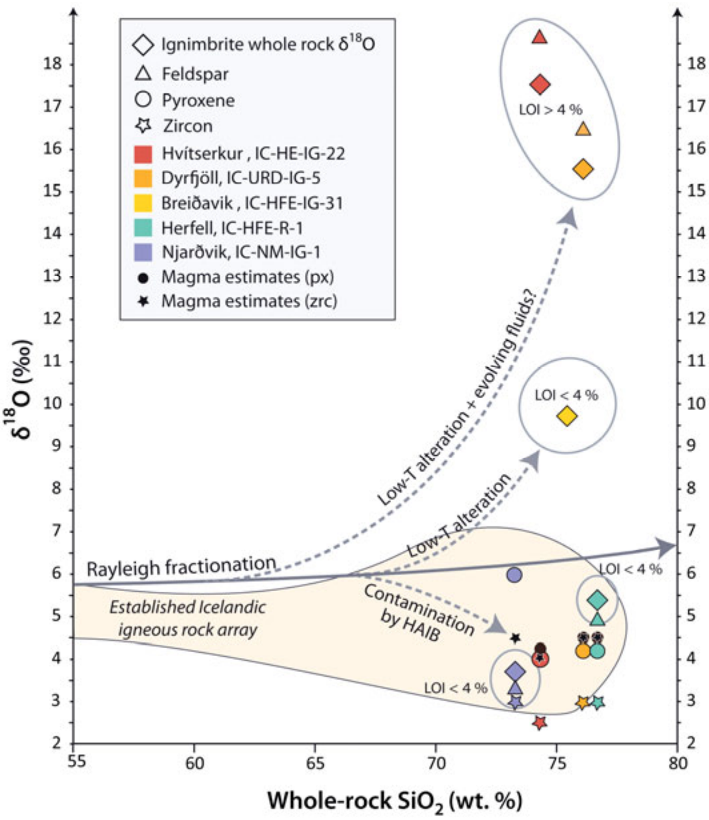

Figure 11. Oxygen isotope values vs. SiO_2 content of the parent whole-rock. Minerals and whole-rocks are plotted with their measured isotopic compositions, and colour coded for each sample. Selected magma estimates from reliable pyroxene (px) and zircon (zrc) minerals are presented with black solid symbols. Zircon data and magma estimates are presented in Paper II. The data show both positive and negative variation around the well-established Raleigh fractionation trend (e.g. Bindeman, 2008), revealing $\delta^{18}O$ contributions from fresh as well as altered constituents in the sampled rock suite with feldspars and some whole-rocks being more susceptible to alteration than pyroxenes (see sample IC-HE-IG-22 and IC-URD-IG-

4.5 Paper V

Floating stones off El Hierro, Canary Islands: xenoliths of pre-island sedimentary origin in the early products of the October 2011 eruption

The submarine eruption that commenced off the south coast of El Hierro, Canary Islands, in October 2011 emitted peculiar eruption products during the first days of activity, which were found floating on the sea surface and drifted for long distances from the eruption site. These decimetre-sized specimens initially resemble lava bombs, but they are characterised by white to cream coloured cores with a porous, pumice-like texture. The nature and origin of the “floating stones” was vigorously debated among researchers, with immediate consequences for hazard assessment of the ongoing eruption.

Paper V, notably the first scientific publication on the “floating stones” and published already during the eruption, presents a detailed textural, mineralogical, elemental and isotopic analysis of several representative samples and compares the result to previous studies on similar rocks found elsewhere in the Canary Islands. The floating stones show prominent vesiculation and degassing textures, high silica content, elevated oxygen isotope values, and a general absence of igneous minerals, while remnant quartz crystals, jasper fragments, carbonate and wollastonite are ubiquitous. These observations lead us to conclude that the early floating stones of El Hierro are vesiculated crustal xenoliths from pre-island sedimentary layers that were entrained and heated by the ascending magma, causing them to partially melt, dehydrate and vesiculate. The textural resemblances of the floating rocks to pumice, together with their xenolithic origin, gave rise to the term “xeno-pumice” for this type of volcanic product. Xeno-pumice provides a window into magma-crust interaction processes and attests that crustal recycling is a relevant process during the early shield-building stage of young edifices. Finally, the occurrence of silica rich xeno-pumice is not, in itself, indicative of explosive high-silica magma beneath El Hierro, but rather demonstrates that partial melting of crustal material generates silica-rich melts.

4.6 Paper VI

Heterogeneous vesiculation of 2011 El Hierro xeno-pumice revealed by synchrotron μ-CT

Paper VI builds on paper V to address the origin and the exact mode of formation of xeno-pumice in further detail, which is relevant because it has bearing on the volcano's eruptive behaviour and therefore also for hazard mitigation. In paper VI, we investigate representative El Hierro xeno-pumice samples through three-dimensional (3D) X-ray computed microtomography (μ-CT) for internal vesicle volumes and vesicle size distributions (VSD) to shed light on their origin. We find a wide range of vesicle sizes and shapes, which are especially variable around small fragments of rock contained in the xeno-pumice samples. Notably, these fragments are almost exclusively of sedimentary derivation, and we interpret these as relicts of the original protolith(s). We consider the heterogeneous vesiculation textures and the irregular VSDs observed to result from pulsed release of volatiles from multiple sources during xeno-pumice formation, which is perhaps best explained by successive vesiculation pulses of pore- and mineral-water from heated sedimentary protoliths (Figure 12). A sedimentary, rather than a magmatic, origin of El Hierro xeno-pumice, by inference reveals that they are products of magma-sediment interaction beneath El Hierro during the course of magma migration through the oceanic crust prior to eruption.

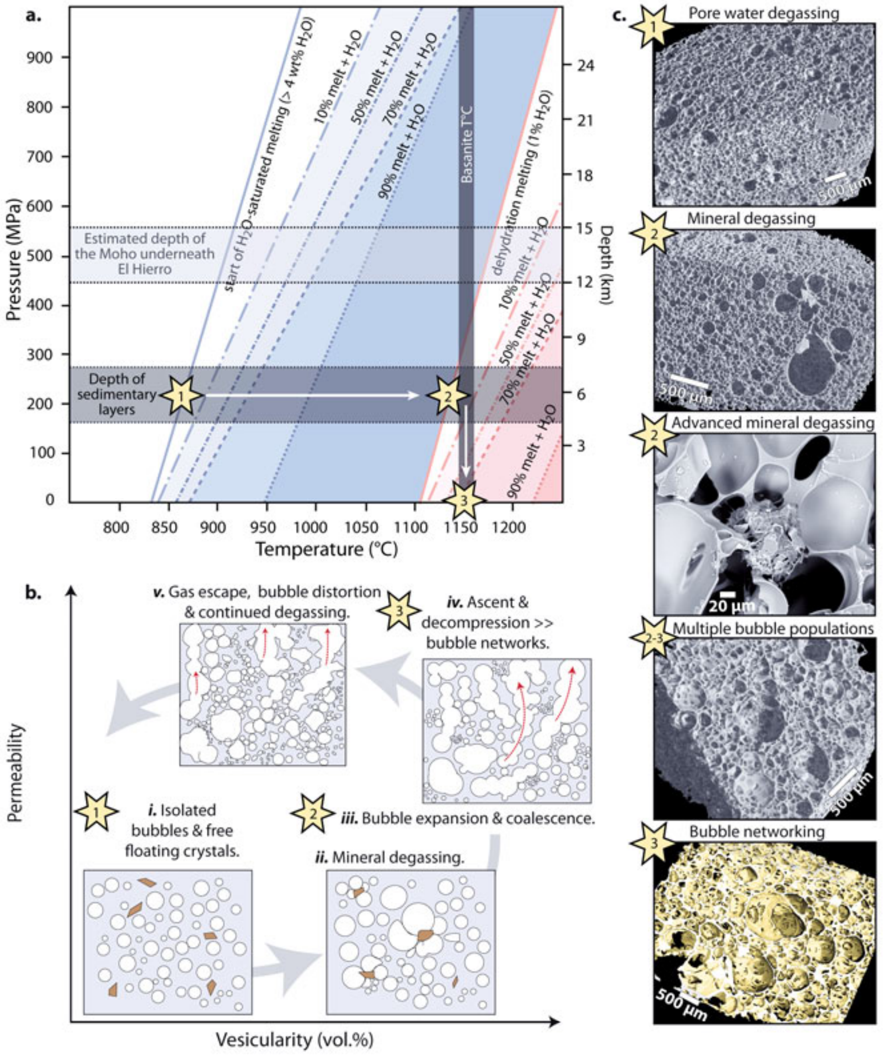

Figure 12. a) Phase diagram reflecting predicted melting behaviour of light coloured El Hierro xeno-pumice calculated by *Perple_X* (modified after Zaczek *et al.*, 2015). The yellow stars denote the pulsed degassing events of pore water during initial H_2O saturated melting (1). Later, liberated mineral water from dehydration melting of mineral phases will also contribute (2). Eventually, the xeno-pumice specimen experienced decompression during ascent in the conduit and associated vesicle-growth, -interaction and -networking (3). b) The pulsed degassing of xeno-pumice is schematically summarised in a vesicularity and permeability hysteresis (cf. Cashman and Sparks, 2013). Steps i) - v) represent key processes during progressive xeno-pumice evolution during degassing and are based on c) snapshots of a degassing sequence as recorded in the associated SEM images and 3D-tomographs.

5. Conclusions and outlook

The outcome of the research presented in this thesis is drawn from the integrated results of textural, petrological, geochemical and in-situ isotope studies, with the purpose to advance our understanding of how silica-rich magma in basalt-dominated oceanic settings can be generated. The first part of the thesis focusses on the disproportionally large volumes of silicic rocks in northeast Iceland and comes to the following conclusions:

- The Borgarfjörður Eystri silicic magmatic cycle in northeast Iceland occurred within a time interval of ≤ 2 Myr and was largely driven by either a flare in plume activity or the formation of a new rift zone, which led to large volume recycling and partial melting of hydrated basaltic crust due to high influx of mantle-derived magma. The geotectonic setting thus appears to have a fundamental control on the generation of silicic magma in plume-related rift environments, where rift relocations into fusible (fertile) crust may trigger rapid production of voluminous mixed origin silicic melt parcels.
- One of the ideal pre-conditions for effective crustal partial melting via basalt injection is an extensively hydrothermally altered crust, which is characteristic for Iceland and has previously been traced via the widespread abundance of low-$\delta^{18}O$ magmatic rocks. However, crustal heterogeneity in $\delta^{18}O$ composition exist among igneous rocks on a local scale, with discrete high-$\delta^{18}O$ compositions recorded e.g. in caldera lake settings, which may become buried under flood basalt lavas within older central volcanoes. The identification of a previously unrecognised high-$\delta^{18}O$ end-member on Iceland opens up the possibility for a wider variety of geochemical end-members to be applied during future assimilation-fractional crystallisation (AFC)-type modelling of Icelandic igneous rocks.
- Oxygen isotope values in zircon from the Borgarfjörður Eystri region fall between 1.2 and 4.5 ‰, and are thus consistently below mantle values, implying that crustal assimilation of ^{18}O-depleted crust was significant in the zircon source. However, the defined crustal heterogeneity with $\delta^{18}O$ compositions up to +18.5 ‰ advocates for a potentially more complex assimilation history involving both ^{18}O-depleted and ^{18}O-enriched crustal rocks.
- Zircons from Borgarfjörður Eystri are dissimilar from average Icelandic zircons with respect to trace elements, REE, and especially Ti-

thermometry temperatures. The northeast Iceland zircons furthermore reveal crystallisation under near water-saturated eutectic melting conditions rather than being products of classical fractional crystallisation. This is consistent with the documented diversity of Icelandic zircons that can be related to the contrasting types of silicic magma generation in rift and off-rift settings in Iceland, which regulates magma temperature, degree of hydration and oxidation state.

- Remarkably, Ti-thermometry temperatures in zircons from Borgarfjörður Eystri are frequently more similar to the values documented for Hadean zircon populations than to some other Icelandic zircon suites. This suggests that these two specific groups of zircon record distinct formation processes, ultimately hinting at crustal recycling as a key process in addition to closed system magma evolution.
- The complex plume-related rift-flank setting that produced the silicic outburst in Neogene northeast Iceland may be analogous to initial continental crust formation, and may thus represent a natural laboratory to better constrain the timescales and magmatic processes required to generate voluminous silicic magma in basalt-dominated settings as recorded for e.g. early Earth.

In the second part of the thesis I address processes active at the magma-crust interface on a much more 'intimate' scale and investigate how crustal materials react upon heating and decompression as they become integrated into basalt-type magma. The textures of crustal xenoliths can provide significant evidence of both xenolith provenance and processes in the magma reservoir and conduit. I target the 2011/12 submarine eruption at El Hierro, an otherwise basalt-dominated ocean island, which brought about pumiceous high-silica inclusions in the basaltic eruption products that were found floating on the sea surface.

- Geochemical investigations and fossil evidence reveal that the high-silica floating pumiceous eruptive products of the El Hierro submarine eruption are most likely xenoliths from pre-island sedimentary layers that became incorporated and heated by the ascending magma. Partial melting and vesiculation of the xenoliths generated pumiceous, high-silica rocks that were coined "xeno-pumice", to characterise their pumice-like texture but foreign provenance to the El Hierro magmatic system.
- The vesiculation history of El Hierro xeno-pumice has been documented via synchrotron based 3D micro-CT and is interpreted to reflect pulsed degassing from a heterogeneous sedimentary protolith. Pore water likely constituted the first and strongest vesiculation pulse, followed by less intense pulses from progressive mineral devolatilisation.
- The xeno-pumice phenomenon is therefore explained as the result of magma-sediment interaction beneath El Hierro prior to eruption. The

vesicle patterns observed reflect intense vesiculation and expansion, and the volatiles released may have increased the volatile load of the magma to temporarily alter the character and intensity of the eruption.

The combined results from both fossil and active volcanic systems in these broadly basalt-dominated oceanic settings provide insights into some key processes of silicic magma generation, and in particular, highlight the interplay between magma and crustal rocks. Importantly, the results underline the timeless relevance of magma-crust interactions from early Earth until today, yet so much is still to be uncovered.

6. Summary in Swedish

Den geokemiska sammansättningen av en primärt basaltisk magma som stiger från manteln upp genom jordskorpan modifieras vanligen av en mängd olika fysiokemiska processer, såsom fraktionerad kristallisation, assimilering av omgivande skorpematerial, blandning av olika magmakroppar, gasutveckling och gasfrigivning. Bildningen av stora mängder kiselrik magma i huvudsakligen basaltiska (kiselfattiga) vulkaniska miljöer är sedan länge ett omdiskuterat problem, och är samtidigt fundamental för vår förståelse av magmatiska processer. Särskilt viktigt är detta för att förstå hur den första kiselrika kontinentala jordskorpan bildades på vår planet för omkring 4 miljarder år sedan. Dessa primära bitar av kontinental jordskorpa på den tidiga jorden, som dominerades av kraftig basaltisk vulkanism, utgjorde de kärnor varifrån dagens kontinenter växte fram och är således en förutsättning livets utveckling. Interaktion mellan magma och skorpematerial är en viktig process i det här sammanhanget eftersom det kan modifiera en magmas geokemiska sammansättning såväl som dess fysikaliska egenskaper.

Den här avhandlingen undersöker hur kiselrik magma bildas på två olika oceaniska vulkanöar. Först undersöks utbrottsprodukter från ett utslocknat vulkanområde omkring samhället Borgarfjörður Eystri på nordöstra Island. Det här området producerade ovanligt stora volymer kiselrika bergarter, vilket avviker från den isländska normen. För att utvidga vår förståelse för dessa vulkaner använder jag flertalet olika analyser av den geokemiska sammansättningen och isotopsignaturer i vulkaniska bergarter samt enskilda zirkonmineral från området. Kombinationen av uran-, bly- och syreisotoper används här för att dels spåra de magmatiska processer som varit verksamma, och dels datera när dessa processer skedde. I avhandlingens andra del studerar jag xenoliter från vulkanutbrottet utanför El Hierro 2011-2012, Kanarieöarna. Xenoliter är främmande bergartsfragment som slitits loss från den omgivande jordskorpan och inneslutits i en framträngande magma på dess väg mot jordytan. Här använder jag mig av geokemiska analyser av xenoliternas sammansättning, samt röntgentomografi av deras 3D-struktur för att förstå interaktionen mellan magma och den omgivande jordskorpan innan vulkanutbrotten.

De studier som genomförts i Borgarfjörður Eystri påvisar att vulkankomplexet, som var aktivt under neogen för omkring 14-12 miljoner år sedan, producerade stora mängder kiselrik magma och efterföljande explosiva vulkanutbrott under en i sammanhanget relativt kort period av mindre än 2 miljoner år. Detta var till följd av en tillfälligt ökad intensitet hos mantelplymen under Island i kombination med att riftzonen försköts, vilket orsakade att stora mängder het magma trängde in i den hydrotermalt omvandlade basaltiska jordskorpan. På grund av den låga smältpunkten hos hydrotermalt omvandlad basalt orsakade magmaintrusionerna partiell uppsmältning av jordskorpan och bildade små fickor av kiselrik magma. Dessa kiselrika magmor ärvde syreisotopsammansättningen av den heterogena moderbergarten. Den isländska jordskorpan är i hög grad sekundärt omvandlad av hydrotermala fluider och kännetecknas generellt av syreisotopvärden som är lägre än vad som är normalt förekommande i manteln, medan syreisotopsammansättningen hos enskilda bergarter lokalt kan variera kraftigt. Detta till trots har bergarter med högre syreisotopvärden än vad som tidigare dokumenterats på Island påträffats under mina undersökningar i Borgarfjörður Eystri, vilket innebär att den geokemiska variationen bland bergarter som kan assimileras i magmareservoarer är större än vad som tidigare antagits.

Island har ofta ansetts vara nutidens motsvarighet till hur den första kiselrika kontinentala jordskorpan bildades på vår Jord under den hadeiska tidsåldern. De zirkonmineral som studerats på nordöstra Island har därför jämförts med de som finns bevarade från hadeikum, och resultaten påvisar flertalet geokemiska likheter mellan de två tektoniska miljöerna. Det innebär således att uppsmältning av jordskorpan i anslutning till nybildade riftzoner troligen var betydelsefull även för bildningen av den första kontinentala jordskorpan.

Den andra delen av den här avhandlingen fokuserar på de besynnerliga och omdiskuterade utbrottsprodukterna som flöt upp till havsytan i början av det flera månaders långa undervattensutbrottet utanför El Hierro som startade i oktober 2011. Geokemiska analyser och jämförelse med olika referensmaterial påvisar att dessa porösa och kiselrika fragment som är omslutna av basaltisk magma i själva verket är krustala xenoliter (dvs. främmande fragment av skorpematerial) som integrerats under magmans passage genom den oceaniska jordskorpan. Dessa xenoliter är kvarvarande rester av de sedimentära lager som finns under den vulkaniska ön och som undkommit total assimilering i magman, medan magmans höga temperatur medfört partiell uppsmältning. De krustala xenoliterna har låg densitet och är extremt bubbelrika, vilket är ett resultat av den betydande mängd volatiler i de sedimentära fragmenten som remobiliserades och frigjordes under partiell smältning och dekompression i det magmatiska systemet. Förutom att assimilering av skorpematerial kan modifiera en magmas geokemiska sammansättning, kan tillförsel av krustala gaser från xenoliterna medföra omfattande konsekvenser för magmans fysikaliska egenskaper. Förändring av en magmas gasinnehåll kan temporärt ändra det följande vulkanutbrottets karaktär och intensitet, med ofta ökad explosivitet till följd.

De resultat som presenteras i den här avhandlingen förbättrar vår förståelse både för processer och tidsramar för hur kiselrik magma bildas i oceaniska miljöer som domineras av basaltisk vulkanism. Den geodynamiska miljön har en signifikant inverkan på de processer som sker och kan vara en kontrollerande faktor som ger unika förutsättningar i varje enskild tektonisk miljö. I synnerhet poängteras att interaktion mellan magma och omgivande skorpmaterial har en betydelsefull inverkan på en magmas geokemiska utveckling, vilket har implikationer för vulkanens utbrottskaraktär och bildandet av kontinental jordskorpa från den tidiga Jorden till idag.

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