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Photograph of a polished slab of foliated peralkaline granite from the Otanmäki area.

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Dr. Asko Käpyaho
Geological Survey of Finland, Espoo

Opponent

Professor Tapani Rämö
University of Helsinki

Petrogenesis of Paleoproterozoic A1-type felsic to intermediate igneous rocks and co-genetic REE-HFSE mineralization in central Finland, Fennoscandian Shield: Evidence from whole-rock geochemistry, Sm-Nd and U-Pb isotope data and thermodynamic modeling

Kimmo Kärenlampi
University of Oulu Graduate School; University of Oulu, Faculty of Technology,
Oulu Mining School, Geosciences
P.O. Box 3000, FI-90014 University of Oulu, Finland

Abstract

The three articles contained in this dissertation deal with the origin, evolution and mineralization of the previously poorly studied suite of 'alkaline granites' in the Otanmäki area, central Finland. These granites, which are assigned to the Otanmäki suite, have drawn intermittent attention from the 1950s to the 1990s due to their partly peralkaline compositions, continental rift-related A-type granite affinity, indications of pre-orogenic age (ca. 1.96–2.05 Ga) and close proximity to the boundary between the Archean Karelia craton and Proterozoic Svecofennian orogen and the Jormua ophiolite complex (1.95 Ga). The Otanmäki suite A-type granites are also economically interesting, because they host two occurrences of rare earth element (REE) and high-field strength element (HFSE) mineralization, which were discovered in the 1980's and contain high abundances of La, Ce, Nd, Y, Zr, Nb, Th and U. Some of these metals, such as the REEs, are essential components used in a wide range of applications, especially in new energy technologies and high-tech consumer products.

The dissertation is based on previous exploration data, new field and drill core observations, petrographic and mineral chemical studies, zircon U-Pb dating, whole-rock major and trace element and Sm-Nd isotope data, and thermodynamically constrained numerical modeling using the Magma Chamber Simulator software. The aim of the study was to 1) resolve the nature and age of the magmatism, 2) make conclusions on the geotectonic environment and petrogenesis of the suite and its relationship to the REE-HFSE mineralization, 3) enhance our understanding of the high-tech metal potential of the Otanmäki suite, and 4) aid future exploration of REE-HFSE resources in Finland.

Field investigations conducted since the beginning of this century have shown that the Otanmäki suite extends over a much larger area than previously thought. During this work, many previously unrecognized rock types were identified in the suite; instead of just 'alkaline granite', a spectrum of peraluminous to peralkaline granites and also intermediate igneous rocks, including syenite and monzodiorite-monzonite, were recognized. These igneous rocks display geochemical characteristics of ferroan A1-type granites typically generated by differentiation of oceanic island basalt (OIB) type magmas in continental rift settings. The original igneous suite was metamorphosed in amphibolite facies conditions and tectonically dismembered into fault-bound thrust sheets (nappes) in Svecofennian tectono-thermal events, being now sliced between Archean tonalite-trondhjemite-granodiorite (TTG) complexes and Paleoproterozoic supracrustal units of the Kainuu belt.

The new geochronological data have clarified the age of the Otanmäki suite magmatism, showing that it took place at ca. 2.04–2.06 Ga. Intermediate to felsic anorogenic plutonic rocks of similar composition and age seem to be missing in other parts of the western margin of the Karelia craton and also in the Fennoscandian Shield as a whole. The general uniqueness of the Otanmäki suite igneous rocks is explained by the interpretation that the nappes containing A1-type rocks represent long-travelled thrust sheets from a more distal, now disappeared part of the western Karelia craton margin.

The Otanmäki suite is a good target for resolving the question of how compositionally diverse bodies of intrusive rocks emplaced in a single suite are formed. The intermediate rocks show relatively juvenile ε_{Nd}(2050 Ma) values (+1.3 to +2.6), which are only slightly lower than the estimated contemporaneous depleted mantle value (+3.4), but much higher than average ε_{Nd}(2050 Ma) of Archean TTGs (–10) in the surrounding bedrock, indicating that these rocks were essentially derived from a mantle source and supporting the view of their derivation by differentiation from a mafic parental magma. The ε_{Nd}(2050 Ma) values of the peralkaline and peraluminous granite samples overlap partly with each other (–0.9 to +0.6 and –3.2 to +0.9, respectively) and are somewhat lower than the values of the intermediate rocks, suggesting that the mafic magmas parental to granite likely have assimilated some amount of older Archean continental crust during their fractionation, which is consistent with the partially continental crust-like incompatible trace element ratios (e.g., Th/Nb, Nb/U) of the granite members.

Based on geochemical and thermodynamic modeling, the origin of the Otanmäki suite igneous rocks is related to differentiation of OIB-like mantle-derived parental magmas by fractional crystallization combined with assimilation of Archean TTG wall rocks, resulting in a spectrum of rocks crystallized from residual melts and melt-cumulus mineral mixtures. These processes took place in the middle to upper crust (~2–4 kbar, ~7–15 km) and involved crystallization of large amounts of clinopyroxene, plagioclase and olivine. The outcomes of the fractional crystallization and assimilation modeling are consistent with previous studies suggesting that magmas of A-type ferroan granites become more peraluminous by involvement of partial melts of quartzo-feldspathic crust, such as Archean TTG gneiss. The modeling also suggests that, in addition to differing amounts of assimilated crustal melts, variations in alkalinity and aluminosity in the end-product granites also depend on the variable fractionation paths of the magmas before the onset of assimilation. In particular, these differences in the parental melt composition, which affect the quantity of late plagioclase fractionation, can explain the variation in the alkalinity and aluminosity of the end-product ferroan A-type granites, even in cases where there was a similar degree of crustal assimilation.

In the Otanmäki area, rocks enriched in REE and HFSE are found in two localities, Kontioaho and Katajakangas. They occur within a peraluminous granite block, which is fault-bound against the adjacent peralkaline granite and syenite. The mineralization is composed of allanite-(Ce)-zircon-titanite-Nb-REE-Th-U oxide assemblages localized in highly fractionated 0.1– to 1.4-m-thick felsic dikes at Katajakangas and a 30– to 50-m-thick felsic sheet-like intrusion at Kontioaho. Allanite-(Ce) is recognized as the major host for REEs in both occurrences. The new results reveal that the REE-HFSE mineralization is genetically unrelated to the peraluminous granite wall rock, but instead, the metal enrichment is intrinsically related to the ca. 2.04–2.05 Ga peralkaline granite magmatism of the Otanmäki suite and represents highly evolved fractionation products of batches of REE-HFSE- and volatile-rich (e.g., F, CO_2, S) peralkaline felsic parental magmas.

Keywords: A1-type granite, ferroan, REE, mineralization, allanite-(Ce), geochemistry, U-Pb dating, Sm-Nd isotopes, Magma Chamber Simulator, continental rifting, Paleoproterozoic, Otanmäki

Keski-Suomessa Otanmäen alueella esiintyvien varhaisproterootsoisten A1-tyypin intermediääristen ja felsisten intrusiivisten magmakivien ja niihin liittyvän REE-HFSE-mineralisaation geokronologia ja synty

Kimmo Kärenlampi
Oulun yliopiston tutkijakoulu, Oulun yliopisto, Teknillinen tiedekunta,
Kaivannaisalan yksikkö, Geotieteet
PL 3000, FI-90014 Oulun yliopisto, Suomi

Tiivistelmä

Tämä väitöskirja sisältää kolme julkaisua, jotka käsittelevät Otanmäen alueella, Keski-Suomessa esiintyvien ’alkaligraniittien’ alkuperää, kehitystä ja mineraalipotentiaalia. Näitä Otanmäen seurueeseen kuuluvia graniitteja on käsitelty muutamissa 1950-1990-luvuille ajoittuvissa tutkimuksissa johtuen niiden A-tyypin graniiteille ominaisista hivenalkuainekoostumuksista sekä preorogeenista syntyä indikoivista ikämääritystuloksista (1,96-2,05 miljardia vuotta), mutta seurueen geologinen merkitys ja alkuperä on jäänyt huonosti ymmärretyksi. Seurueen graniitit esiintyvät lähellä arkeeisen karjalaisen kratonin ja paleoproterosooisen svekofennisen orogeenisen vyöhykkeen rajaa ja myös lähellä 1.95 miljardin vuoden ikäistä Jormua ofioliittikompleksia. Otanmäen seurueen graniitit ovat myös taloudellisessa mielessä mielenkiintoisia, sillä niistä tunnetaan kaksi mineralisoitunutta vyöhykettä (Katajakangas ja Kontioaho), jotka sisältävät korkeita pitoisuuksia harvinaisia maametalleja (engl., rare earth element, REE), kuten lantaania (La), ceriumia (Ce), neodyymiä (Nd) ja yttriumia (Y), sekä korkean kenttävoimakkuuden alkuaineita (engl., high-field strength elements, HFSE), kuten zirkoniumia (Zr), niobia (Nb), toriumia (Th) ja uraania (U). Osa näistä metalleista on ns. high-tech -metalleja, joita käytetään laajasti monissa korkean teknologian käyttökohteissa.

Tässä väitöskirjatyössä tutkitaan Otanmäen seurueen kivilajeja ja niiden alueellista esiintymistä käyttäen kenttä- ja kairasydänhavaintoja, petrografiaa ja mineraalikemiallisia määrityksiä, zirkonin U-Pb-ikämääritystuloksia, kokokivien pää- ja hivenalkuainekoostumuksia ja Sm-Nd-isotooppianalyysejä sekä termodynaamista numeerista mallinnusta Magma Chamber Simulator -mallinnusohjelmistolla. Tutkimuksen tavoitteena on selvittää Otanmäen seurueen magmatismin kemiallisia ominaispiirteitä, geotektonista ympäristöstä ja alkuperää. Lisäksi tutkimuksessa arvioidaan seurueen ja REE-HFSE mineralisaation geneettistä suhdetta ja sen high-tech -metallipotentiaalia. Uutta tietoa voidaan käyttää malminetsinnän lähtöaineistona.

Tämän vuosituhannen alussa aloitetut kenttätutkimukset ovat osoittaneet, että Otanmäen seurueen esiintymisalue on laajempi kuin aikaisemmin luultiin. Tässä tutkimuksessa osoitetaan, että se sisältää korkean Fe/Mg-suhteen omaavia, peralumiinisia ja peralkalisia graniitteja ja lisäksi siinä on aikaisemmin tunnistamatta jääneitä intermediäärisiä magmakiviä, kuten syeniittiä ja montsodioriittia-montsoniittia. Otanmäen seurueen magmakivet muistuttavat geokemiallisilta ominaispiirteiltään mantereisten repeämäympäristöjen A1-tyypin graniitteja, joiden alkuperä yleensä liitetään koostumukseltaan merellisten saarten basaltteja muistuttavien vaippaperäisten sulien differentioitumiseen. Seurueen kivet metamorfoituivat amfiboliittifasieksen olosuhteissa svekofenniseen orogeniaan liittyvissä tektonotermisissä tapahtumissa ja pilkkoutuivat tektonisesti ylityöntölaatoiksi (nappe), jotka rajautuvat siirroksilla arkeeisiin tonaliitti-trondhjemiitti-granodioriittikomplekseihin (TTG-komplekseihin) ja Kainuun liuskejakson paleoproterosooisiin suprakrustisiin yksiköihin.

Uudet ikämääritykset ovat tarkentaneet aiemmissa tutkimuksissa epäselväksi jäänyttä Otanmäen seurueen magmatismin ikää ja osoittaneet, että magmatismi tapahtui 2,04-2,06 miljardia vuotta sitten. Samanlaisen koostumuksen ja iän omaavia kiviä ei tunneta muualta Karjalan kratonin länsiosista eikä koko Fennoskandian kilpialueelta. A1-tyypin kivien harvinaisuutta selittää tulkinta, jonka mukaan ylityöntölaatat ovat työntyneet nykyiseen asemaansa kaukaisesta, svekofennisen orogenian alkuvaiheessa poisleikatusta Karjalan kratonin läntisestä reunaosasta.

Otanmäen seurue on hyvä kohde tarkastella magmaattista systeemiä, joka sisältää koostumukseltaan laajasti vaihtelevan valikoiman erilaisia A-tyypin kiviä. Seurueen intermediääristen kivien initiaaliset ε_{Nd}-arvot ovat juveniilisia (+1.3-+2.6) ja vain vähän matalampia kuin samanaikaisella köyhtyneellä vaipalla arvioidaan olleen, mutta paljon korkeampia kuin ympäröivän kallioperän arkeeisilla TTG-gneisseillä (ka. –10), mikä tukee tulkintaa seurueen intermediääristen kivien synnystä vaippaperäisen mafisen kantasulan differentoitumisen kautta. Seurueen peralkalisten ja peralumiinisten graniittien osin päällekkäiset ε_{Nd}(2050 Ma)-arvot (–0.9-+0.6 ja –3.2-+0.9) ovat alhaisempia kuin intermediäärisillä kivillä, mikä osoittaa, että graniittien mafiset kantasulat todennäköisesti ovat assimiloineet arkeeista kuorta. Tätä tulkintaa tukevat myös graniittien keskimääräistä mantereista yläkuorta ja arkeeisia TTG-gneissejä muistuttavat Nb/U- ja Th/Nb-suhteet.

Geokemialliseen ja termodynaamiseen mallinnukseen perustuen Otanmäen seurueen kivilajikirjo edustaa kumulusmineraalien ja jäännössulien seoksia ja jäännössulista kiteytyneitä kiviä, jotka syntyivät valtamerten saarten basaltteja muistuttavista vaippaperäisistä mafisista kantasulista fraktioivan kiteytymisen ja samanaikaisen arkeeisten TTG-gneissien assimiloinnin kautta. Differentaatioprosessit tapahtuivat ylä- ja keskikuoren olosuhteissa (~2-4 kbar, ~7-15 km), ja niiden seurauksena on täytynyt syntyä suuria määriä mafisia kumulaatteja. Fraktioivan kiteytymisen ja assimilaation mallinnuksen tulokset tukevat, paitsi aikaisempia käsityksiä korkean Fe/Mg-suhteen omaavien A-tyypin graniittien magmojen synnystä, myös sitä olettamusta, että niiden koostumukset muuttuvat peralumiinisemmiksi assimiloimalla mantereisen kuoren kvartsi- ja maasälpärikkaista kivistä, kuten arkeeisista TTG gneisseistä peräisin olevia osittaissulia. Mallinnus osoittaa myös, että erot kantasulien koostumuksissa vaikuttavat myöhäisessä kehitysvaiheessa kiteytyvän plagioklaasin määrään, mikä selittää graniittien alkalisuuden ja alumiinisuuden vaihteluita myös sellaisissa tapauksissa, joissa assimiloidun mantereisen kuoren määrät ovat samansuuruisia.

Katajakankaan ja Kontioahon REE-HFSE-mineralisoituneet vyöhykkeet esiintyvät siirrosten rajaamassa lohkossa, joka koostuu lähinnä Otanmäen seurueen peralumiinisesta graniitista (2,06 miljardia vuotta) ja rajautuu siirroksin Otanmäen seurueen peralkaliseen graniittiin ja syeniittiin. Katajakankaalla REE-HFSE-rikastumat sijoittuvat isäntägraniittiin tunkeutuneisiin, terävästi rajautuneisiin ja 0,1-1,4 metrin paksuisiin felsisiin juoniin ja Kontioahossa 30-50 metriä paksuun, levymäiseen intruusioon. Ne sisältävät Ce-rikasta allaniittia, zirkonia, titaniittia ja erilaisia Nb-REE-Th-U-oksideja. Allaniitti-(Ce) on merkittävin lantanidien kantajamineraali molemmissa mineralisoituneissa vyöhykkeissä. REE-HFSE-mineralisaatio liittyy läheisesti Otanmäen seurueen noin 2,04-2,05 miljardin vuoden ikäiseen peralkaliseen magmatismiin ja mineralisoituneet juonet ja intruusio edustavat REE-HFSE- ja volatiilirikkaiden (esim. F, CO_2 ja S) peralkalisten felsisten magmojen pitkälle kehittyneitä lopputuotteita.

Asiasanat: A1-tyypin graniitti, REE, mineralisaatio, allaniitti-(Ce), geokemia, ikämääritys, U-Pb isotoopit, Sm-Nd isotoopit, Magma Chamber Simulator, mantereinen repeytyminen, paleoproterootsoinen, Otanmäki

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I express my sincere gratitude to my principal supervisor, Prof Eero Hanski, as I would not have even considered starting a PhD project if he had not encouraged me to do so and for his persistence in teaching me academic and scientific skills. This thesis benefitted greatly from the pioneering work of Asko Kontinen on the geology of the Kainuu area and his inspirational attitude and openness to share his knowledge. Asko was engaged in numerous discussions during writing of the manuscripts and interpretation of the data and occasionally stepped in as a supervisor, when my ideas started to get too wild. Hannu Huhma, Yann Lahaye, and Hugh O'Brien from the Finnish Isotope Geosciences laboratory, Geological Survey of Finland, are thanked for their expert guidance in isotope analysis and data interpretation. Jussi S. Heinonen is thanked for his encouraging attitude and guidance with the Magma Chamber Simulator, as without his expertise, I believe I would still be sitting at my computer doing AFC modeling. Dr. Joachim Krause and Thomas Heinig, technicians and other staff of the Helmholtz-Zentrum Dresden-Rossendorf are thanked for their assistance, guidance and hospitality during my visit in Freiberg. I also want to express my gratitude to Kari Strand, Sheng-Hong Yang, Sari Forss, Riitta Kontio, Jukka Laurikkala, and other staff of OMS who supported me during my project. Perttu Mikkola, Eija Vallimies, and Vesa Nykänen are thanked for assisting in getting access to GTK data archives and Marko Puonti for assistance and hospitality at the National Drill Core Archive at Loppi. Jukka-Pekka Ranta, Kirsi Luolavirta, Marko Moilanen, and other PhD students at OMS are thanked for their support along the way. Anssi Mäkisalo, Jorma Mikkonen, and Juho Tapio are thanked for assistance in the field work and sampling. I also present special thanks to Jouko Jylänki, CEO of the Otanmäki Mine Oy, for all his encouragement and endless optimism during the project. I am grateful to my family, Laura and Lumi for all support.

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Oulu, 18.05.2021

Kimmo Kärenlampi

List of the original publications

The original papers on which this thesis is based are referred to in the text by the Roman numerals I–III.

I: Kärenlampi., K., Kontinen, A., Huhma, H., Hanski, E., 2019. Geology, geochronology and geochemistry of the 2.05 Ga gneissic A1-type granites and related intermediate rocks in central Finland: implication for the tectonic evolution of the Karelia craton margin. Bulletin of the Geological Society of Finland 91, 35–73.

II: Kärenlampi, K., Heinonen, J.S., Kontinen, A., Hanski, E., Huhma H., 2021. Geochemical and thermodynamic modeling of the petrogenesis of A1-type granites and associated intermediate rocks: a case study from the central Fennoscandian Shield. Geochemistry 81, 125734.

III: Kärenlampi, K., Kontinen, A., Hanski, E., Huhma, H., Lahaye, Y, Krause, J., Heinig, T., 2020. Age and origin of the Nb-Zr-REE mineralization in the Paleoproterozoic A1-type granitoids at Otanmäki, central Finland. Bulletin of the Geological Society of Finland 92, 39–71.

The contributions of the first author and the co-authors in the original publications.

I: K. Kärenlampi had the main responsibility for the manuscript preparation and interpretation of the results. A. Kontinen contributed to the manuscript preparation, especially to the interpretation of the tectonic setting of the Otanmäki suite. H. Huhma was responsible for the ID-TIMS and LA-MC-ICP-MS U-Pb analysis and sample preparation and contributed to the interpretation of the U-Pb data. E. Hanski contributed in supervision and manuscript preparation.

II: The main responsibility for the manuscript preparation and interpretation was carried by K. Kärenlampi with other authors contributing. K. Kärenlampi was responsible of the Magma Chamber Simulator (MCS) thermodynamic modeling and interpretation with J.S. Heinonen contributing to the modeling and interpretation of the modeling results. A. Kontinen and E. Hanski contributed to the supervision, preparation and reviewing the manuscript drafts. H. Huhma was responsible for the Sm-Nd isotope analysis and contributed to the interpretation of the Sm-Nd data.

III: K. Kärenlampi was responsible of sampling, interpretation and preparing the manuscript. A. Kontinen and E. Hanski contributed by reviewing the manuscript drafts. H. Huhma and Y. Lahaye were responsible for the Sm-Nd isotope analysis and sample preparation. H. Huhma also contributed to the interpretation U-Pb and Sm-Nd data. J. Krause was responsible of the electron microprobe analyses and T. Heinig for the FE-SEM based MLA scans in co-operation with K. Kärenlampi.

Contents

1 Introduction

The Proterozoic granitoid magmatism in Finland is dominated by widespread Svecofennian pre-, syn- and post-orogenic plutonic magmatism, which took place in the time interval of 1.77–1.93 Ga (Nironen, 2005), and subsequent 1.54–1.64 Ga anorogenic rapakivi granites (Rämö and Haapala, 2005). Before 1.93 Ga, the Paleoproterozoic geological evolution only involved rare felsic intrusions and was dominated by deposition of Karelian sedimentary and mafic and minor felsic volcanic rocks on the Archean basement and emplacement of coeval mafic intrusions (Laajoki, 2005; Iljina and Hanski, 2005; Vuollo and Huhma, 2005; Hanski, 2013; Hanski and Melezhik, 2013a, b).

Generally, the post-Archean felsic intrusions found in northern, central and eastern Finland are relatively small granitic plutons, which have ages between 2.4 and 2.0 Ga (Ahtonen et al., 2007; Mikkola, 2011; Ranta et al., 2015). They also include the gneissic ‘alkaline granites’ that were recognized in the Otanmäki area (Fig. 1) in the 1950s and 1960s based on their high total alkali and FeO_{tot} contents, high Fe/Mg ratios and elevated rare earth element (REE), high-field strength element (HFSE) contents and the presence of alkali pyroxene and amphibole (Pääkkönen, 1956; Marmo et al., 1966). These granites (the Otanmäki suite) have drawn attention due to their close proximity to the western margin of the Archean Karelia craton and the 1.95 Ga Jormua ophiolite complex, with the latter representing ancient seafloor and their chemical affinity to A-type granites related to continental rifting (Peltonen et al., 1996, 1998; Kontinen et al., 2013a). Against this background, the Otanmäki suite is clearly an important piece in the puzzle of understanding the Paleoproterozoic evolution of the western margin of Karelia craton. However, published information has remained minimal and the spatial distribution, geology, origin and geotectonic significance of the Otanmäki suite rocks have remained poorly understood. In addition, previous studies report confusingly variable U-Pb zircon ages ranging from ca. 1.96 Ga to 2.05 Ga (Marmo et al., 1966; Hytönen and Hautala, 1985; Peltonen et al., 1996; Kontinen et al., 2013a).

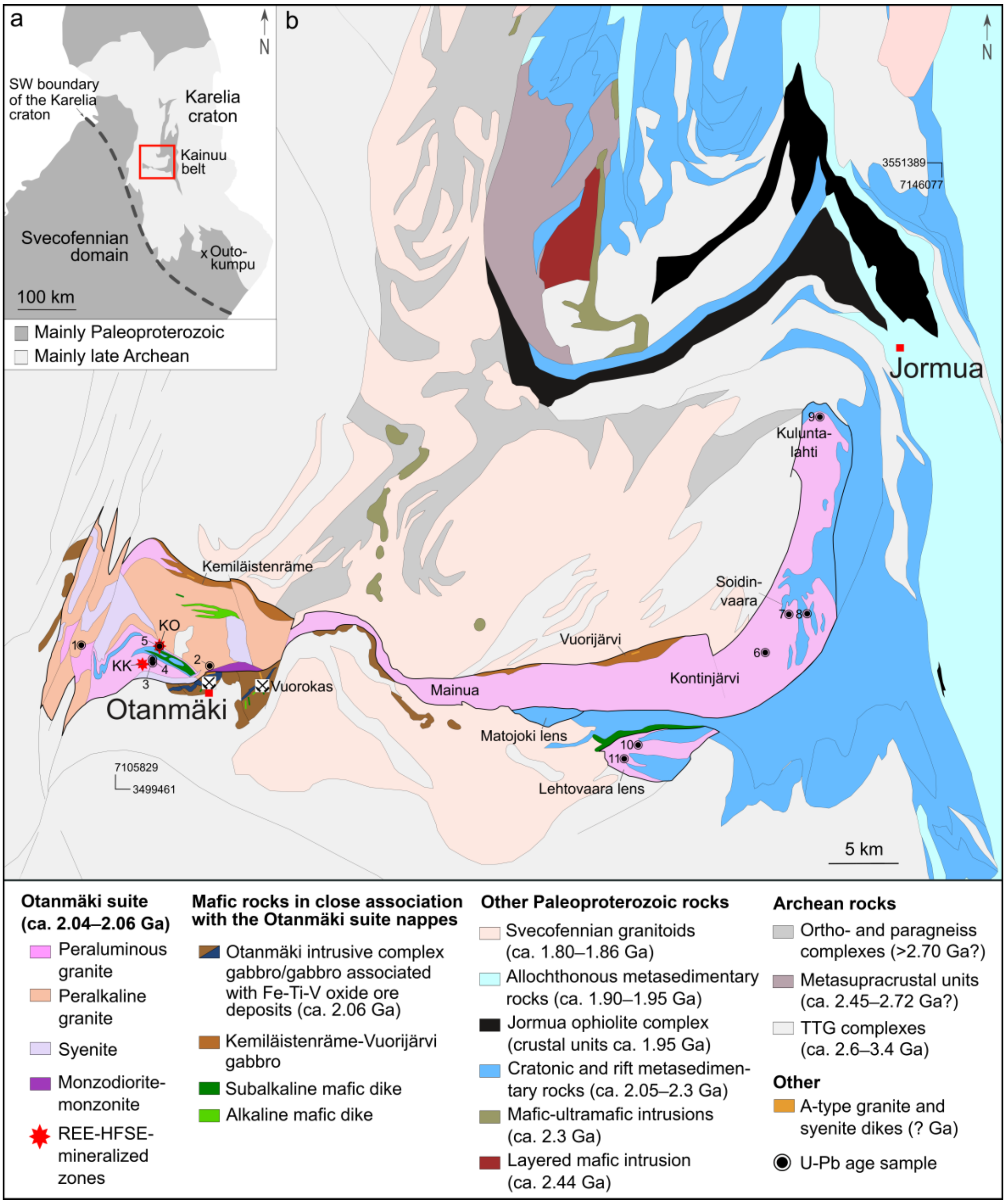

Figure 1. a) Location of the study area (red rectangle) close to the SW margin of the Archean Karelia craton. b) Map showing the geological setting of the Otanmäki suite A1-type felsic to intermediate igneous rocks. Also shown are the locations (black circles) of the U-Pb zircon dating samples (see Table 1) and the closed Otanmäki and Vuorokas Fe-Ti-V mines. KO = Kontioaho and KK = Katajakangas. Modified after Bedrock of Finland – DigiKP and Papers I-III. Coordinates in KKJ-3/Finland Uniform Coordinate System.

The Otanmäki suite A-type granites are also economically of special interest, as they are associated with significant REE and HFSE enrichments. The first observations of REE mineralization in the Otanmäki area were made in 1981 from radioactive, U-Th-bearing glacial boulders having elevated concentrations of REEs and HFSEs, such as Nb and Zr (Äikäs, 1990). The follow-up exploration drilling program of the Rautaruukki Oy mining company between

1983 and 1985 led to the discovery of two REE-HFSE-mineralized zones in bedrock, which were named Katajakangas and Kontioaho (Hugg and Heiskanen, 1986). According to the preliminary mineral resource estimates from that time, the Katajakangas mineralization contains 0.46 Mt of rock with 2.3 wt% total rare earth elements (TREE), 0.5 wt% Nb, and 0.8 wt% Zr (Hugg, 1985a), and the Kontioaho mineralization contains 4 Mt of rock with 0.6 wt% TREE, 0.08 wt% Nb, and 2.1 wt% Zr (Hugg, 1985b). These estimates were based on sparse drilling information and the depth and strike extensions of the mineralized zones remained unconstrained. In 1985, Rautaruukki Oy discontinued its REE project and concluded that the identified resources are either too small (Katajakangas) or the grade is too low (Kontioaho) to be economically exploitable taking in account the price of REE at that time (Hugg and Heiskanen, 1986). The decision to terminate the Otanmäki REE project was made at the same time as the company's decision to close all its exploration activity in Finland and also the Otanmäki Fe-Ti-V mine.

Although the Otanmäki REE occurrences have been known already since the 1980s and despite the large amount of data generated during their exploration, systematic research and published information on the mineralization have remained minimal. In recent years, the demand for REE has dramatically increased as they have become increasingly important due to their applications in a variety of high-tech devices (e.g., hybrid and electric vehicles, wind turbine generators, compact fluorescent lights, flat screen televisions, mobile phones, disc drives, defense technologies etc.) and chemical processing (Chen and Zheng, 2019; Coeya, 2020; USGS, 2021). In the European Union, the REEs are included in the list of critical raw materials (EU, 2020), which are materials that have a significant economic importance, a high supply risk and presently a lack of reasonable substitutes. There is currently no mining of REEs in the EU as all REE raw materials or intermediate products are imported from Australia, USA, India, and China (ERECON, 2015; Machachek and Kalvig, 2016; EURARE, 2017, 2018; EU, 2020; USGS, 2021). Due to the EU's high dependence on the importation of REE, much attention has recently been paid to discovering new mineral resources within the EU. One of these efforts includes the High-Tech Metal Project of the Geological Survey of Finland (GTK) (Sarapää et al., 2015), the aim of which was to assess the high-tech metal potential of Finland. This evaluation listed the Otanmäki REE-HFSE occurrences among the most promising high-tech metal targets in Finland.

Based on the above-discussed background, the Otanmäki suite granites and related REE-HFSE occurrences constitute a highly interesting and many ways relevant research subject for a PhD project. In the first place, the suite provides a highly interesting target deemed to carry significant information on the timing and geological processes related to the break-up of the Karelia craton and the petrogenesis of a suite of A-type granites in the Otanmäki area and A-type granites in general. Moreover, the REE-HFSE mineralization is seen as a good target to study REE-HFSE-mineralizing processes and further our understanding on the high-tech metal potential of the Otanmäki suite, which have not been properly discussed in the literature prior to Papers I-III. The decision to start the project was also influenced by the existence of a considerable amount of unpublished data on the Otanmäki suite granites and REE-HFSE mineralization, available from GTK, Rautaruukki Oy and Otanmäki Mine Oy.

The synopsis is divided into three parts. The first part presents a summary of the state of the art in research of A-type granites and related REE-HFSE mineralization. Also, a review of the regional geological setting of the Otanmäki area is given and the results of previous studies on the Otanmäki suite and its REE-HFSE mineralization prior to this research are summarized. The second part concerns key research results presented in the three publications and answers to the main research questions. The third part of the synopsis discusses what new this research tells us in the context of the geological evolution of the Karelia craton, petrogenesis of A-type granites and origin of REE-HFSE mineralization in general.

2 State of the art in research of A-type granites and related REE-HFSE mineralization

2.1 General background

The A-type (‘A’ variously defined as alkaline, anorogenic, anhydrous, or ambiguous; e.g., Bonin, 2007) granites are rather enigmatic rock types, not only due to uncertainties regarding their typology and petrogenesis, but also in terms of their tectonic setting and overall significance in the evolution of Earth’s lithosphere. They show different modes of occurrence, typically ranging from plutonic complexes and batholiths in the middle to upper crust to subvolcanic and volcanic complexes (Bonin, 2007; Frost and Frost, 2013). They often occur in association with coeval mafic to intermediate rocks, such as anorthosite-gabbro-diorite, monzogabbro-monzodiorite-monzonite, and/or quartz syenite-alkali feldspar syenite, although most suites only contain a portion of this compositional spectrum (see Bonin, 2007; Frost and Frost, 2013 for overview). These suites are typically characterized by three lithologic associations: 1) felsic intrusions related to contemporaneous large layered mafic intrusions, 2) felsic or intermediate intrusions emplaced in post- or late-collisional settings, and 3) felsic or intermediate intrusions in rift settings (Eby, 1992; Bonin, 2007; Frost and Frost, 2013). Globally, suites of A-type granites and related mafic to intermediate rocks (and their volcanic equivalents) span almost three billion years from late Archean (ca. 2.8 Ga) to Phanerozoic (Bonin, 2007 and references therein; Dall'Agnol et al., 2012; Frost and Frost, 2013; Grebennikov, 2014).

The petrogenesis of A-type granites has been the subject of extensive research over the last 40 years and proposed models on their origin have been constantly evolving (e.g., Bonin, 2007; Dall'Agnol et al., 2012). The most frequently suggested petrogenetic models propose that A-type granites can originate both through differentiation of silicate magmas and partial melting of continental crust in a variety of tectonic settings (e.g., Frost and Frost, 2011). Later researchers have been working towards unifying the incoherent terminology used in describing A-type igneous suites (Bonin, 2007; Frost and Frost, 2011; 2013; Dall'Agnol et al., 2012; Bonin et al., 2019).

The A-type granites are also of significance due to their potential of hosting economic resources of REE and HFSE (e.g., Zr, Nb, U, Th) and other metals, such as Cu, Au, Sn, and W (Bonin, 2007; Costi et al., 2009; Dall'Agnol et al., 2012; Dostal, 2016; Vasyukova and Williams-Jones, 2020). There has been much recent interest from the exploration industry and researchers to A-type granites and related deposits due to the rise of the prices of REEs (Dostal, 2016). Recent studies (e.g., Kynicky et al., 2011; Kempe et al., 2015; Dostal et al., 2014; Vasyukova and Williams-Jones, 2014, 2020) have shown that hyper-enrichment of REE and HFSE to potentially economically exploitable degrees in A-type granites requires a combination of magmatic and magmatic-hydrothermal processes.

2.2 A-type granites and related rocks: their classification and compositional variability

Granite (*sensu stricto*) is a plutonic rock with a simple felsic mineral assembly of quartz + alkali feldspar + plagioclase, in which the quartz content is more than 20 vol% and plagioclase makes 10 to 65 vol% of total feldspar (Le Maitre et al., 2002). Despite their apparently simple basic mineralogical compositions, granites are widely varied in terms of their modal and chemical compositions and modes of origin and occurrence (Brown, 2013; Bonin et al., 2019). Several classification schemes have been introduced to discriminate between various types of granite. The current schemes used for classification/naming of granites are based on their modal mineralogy or whole-rock key chemical parameters, of which most significant are aluminosity, alkalinity and the balance between Fe and Mg (Frost et al., 2001; Bonin et al., 2019).

Aluminosity and alkalinity, which are typically expressed as A/CNK (molecular ratio of $Al_2O_3/(CaO+Na_2O+K_2O)$) and A/NK (molecular ratio of $Al_2O_3/(Na_2O+K_2O)$, respectively, have long been regarded as the key properties of granites as they reflect the excess or deficit of Al relative to feldspar. Based on the aluminosity criterion, peralkaline (A/NK<1, A/CNK<1), metaluminous (A/NK>1, A/CNK<1) and peraluminous (A/NK>1, A/CNK>1) granite varieties are recognized. The granites containing an excess of alumina crystallize Al-excess minerals (e.g., aluminosilicates, such as biotite, muscovite, garnet) in addition to feldspar whereas those that are Al-deficient crystallize Al-deficient minerals (e.g., amphiboles, pyroxenes). A more recent classification of granites focuses on the balance between Fe and Mg, or Fe* or the Fe - number (Frost et al., 2001), calculated as weight proportions of $FeO_{tot}/(FeO_{tot}+MgO)$, which distinguishes granites into magnesian (Fe* = <0.8) and ferroan (Fe* = >0.8) types.

The petrogenetic granite classifications schemes rely on the combination of petrographical and chemical data, including trace elements and isotopes (see a review in Bonin et al., 2019). One of the oldest of such approaches and still widely used is the alphabetic S-I-M-A genetic classification. It was presented originally by Chappell and White (1974) and has been elaborated over the years (e.g., Chappell 1999; Chappell and White, 2001; Collins et al., 2019). The alphabetic classification recognizes contrasting suites of granites whose major element compositions are assumed to reflect their sources. These suites are: 1) S-type granites, which are relatively potassic, peraluminous and high-SiO_2 granites, presumably originated via partial melting of metasedimentary source rocks; 2) I-type granites (I = igneous), which are sodic, metaluminous, mafic to felsic granites thought have originated by partial melting of meta-igneous rocks; 3) M-type granites (M = mantle-derived sources) derived by melting of subducted oceanic crust or the overlying mantle wedge at active continental margins or by differentiation of mantle-derived mafic magmas at oceanic islands or mid-ocean ridges; 4) A-type granites generated typically along rift zones and in stable intracontinental settings. The A-type granites are distinguished from the I- and S-types by their elevated contents of alkalies, FeO_{tot}, F, HFSE (Zr, Nb, Ta) and REE (except Eu), as well as high Fe/Mg and Ga/Al ratios and typically low concentrations of CaO and trace elements compatible with mafic silicates (Cr, Ni, Co, and Sc) and feldspars (Ba, Sr, and Eu). The letter ‘A’ referred originally to an anorogenic setting but its meaning has become less clear in modern petrology (see below). Differences in compositions of the alphabetic S-I-A suites are reflected in their distinct modal mineralogy. Peraluminousity of S-type granites is demonstrated by a mineral assemblage containing muscovite, cordierite, aluminosilicates and monazite, whereas the presence of hornblende and titanite in I-type granites indicates a metaluminous character. The chemistry of A-type granites is reflected in the occurrence of characteristic Fe-rich mafic silicates (fayalite, annite, or hastingsite), including alkali pyroxenes and/or amphiboles (aegirine-augite, aegirine, arfvedsonite, riebeckite) in peralkaline rocks.

The first introduction of the term ‘A-type granite’ is attributed to Loiselle and Wones (1979), who used the term in a conference abstract to describe a previously unrecognized group of iron-rich granitoids that occur in intracontinental settings. The original definition of the term ‘A-type’ referred to mildly alkaline (peralkaline) granites with high Fe/Mg and elevated HFSE (e.g., Zr, Nb, Ta) and REE (except Eu) contents and low contents of Co, Ni, Sc, Cr, Ba, Sr and Eu. Subsequently, the term acquired a much broader definition. The first use of the A-type nomenclature in a research paper is found in Collins et al. (1982), who assigned granite complexes of southeastern Australia to the A-type based on their ferroan composition and high contents of alkalis, REE, Nb and Ga. However, the A-type granites in Collins et al. (1982) included not only peralkaline rocks but also peraluminous to metaluminous rocks.

Later, the meaning of the term ‘A-type’ became widened to include even a greater diversity of rocks, especially after Pearce et al. (1984) and Whalen et al. (1987) introduced their tectonomagmatic granite discrimination diagrams, in which A-type granites are discriminated based on their distinctive REE-HFSE and LILE characteristics. Subsequently, A-type granites were recognized from various tectonic settings corresponding not solely to the anorogenic regime but also environments at the end of an orogenic cycle in late collisional to post-orogenic stages, and their origins appeared to differ widely (e.g., White and Chappell, 1983; Brown et al., 1984; Capaldi et al., 1987; Sylvester, 1989; Kleeman and Twist, 1989; Maniar and Piccoli, 1989; Rogers and Greenberg, 1990; Eby 1990, Creaser et al., 1991; Rämö, 1991; Coleman et al., 1992). A great deal of confusion arose when granites with different compositions and likely differing origins were combined in the same ‘type’, making the usage of the term ‘A-type’ ambiguous. Eby (1992) drew an influential conclusion that A-type granites may be produced in different ways and also in other than anorogenic regimes and hence their discrimination should be based on their geochemical properties rather than their geodynamic settings. Eby (1992) suggested a subdivision into two major types based on abundances of trace elements, such as Y, Nb, Ce, Ga and, in particular, Y/Nb. Granites with lower Y/Nb ratios (A1-type, Y/Nb <1.2) include felsic igneous rocks from oceanic islands and continental rifts with a chemical affinity to OIBs. Granites with higher Y/Nb (A2-type) are thought to correspond to a wider range of geodynamic settings and granite compositions, including felsic intrusions related to contemporaneous layered mafic intrusions and felsic or intermediate intrusions emplaced in post- or late-collisional settings. However, the data set used by Eby (1992) also illustrated that some A1-type suites display a gradual shift from within-plate A1 to post-collision A2 compositions, showing that there is some uncertainty in the classification at the A1–A2 boundary.

Frost et al. (2001) reasoned that classification of granitic rocks should be based on major elements, because trace elements in granitic rocks are commonly not incompatible and therefore their abundances and ratios in granites cannot be unambiguously used to identify magma sources or tectonic settings. Accordingly, Frost et al. (2001) proposed a specific classification of felsic igneous rocks using four chemical parameters: 1) ferroan index $FeO_{tot}/(FeO_{tot}+MgO)$ (ferroan vs. magnesian); 2) modified alkali–lime index ($Na_2O+K_2O–CaO$) (MALI) discriminating between alkalic, alkali-calcic, calc-alkalic and calcic; 3) alumina saturation index (ASI) (molecular Al/(Ca–1.67×P+Na+K); and 4) alkalinity index (AI) (molecular Al–(Na+K)). Using these indexes of Frost et al. (2001), Frost and Frost (2011) divided A-type granites into four geochemical groups of different origin and evolution and observed that their generation is not restricted to any specific tectonic setting. These include: 1) alkalic granites that may be metaluminous or peralkaline, 2) alkali-calcic granites that may be metaluminous, peraluminous or peralkaline, 3) calc-alkalic granites that may be metaluminous or peraluminous, and 4) rare calcic metaluminous ferroan granites. This classification scheme was a step forward as it demonstrated that 1) the geodynamic setting of A-type granite magmas cannot be decisively discriminated solely on the basis of chemical characteristics, and 2) in ambiguous cases, the relevance of any geochemistry-based geodynamic/geotectonic classification scheme of A-type granites should be carefully tested using all available relevant geological, geochemical and isotopic data.

2.3 Origin of A-type granites

Several modes of origin have been proposed for A-type granites, which is not surprising given the varying compositions of the granitic rocks considered to be of the ‘A-type’. Loiselle and Wones (1979) first proposed that A-type granites included in their data set may have formed by fractionation of mantle-derived alkali basalt, with or without crustal contamination involving depleted granulite facies lower crustal rocks. Collins et al. (1982) proposed that vapor-absent melting of depleted lower crustal rocks alone could produce A-type granite magmas. However, the ‘residual source model’ did not receive widespread acceptance. For example, Creaser et al.

(1991) presented that partial melts from such a source should display major element characteristics opposite to those shown by most A-type granites. Instead, they argued that the major element compositions of most A-type granites could be produced by 15% to 40% partial melting of tonalitic and granodioritic meta-igneous source rocks. A similar model was earlier proposed by Anderson (1983) who suggested that major element compositions of voluminous anorogenic rapakivi granites in the Mid-Continental United States likely were derived from melting of tonalite-granodiorite-diorite source rocks. Experimental results supported these propositions by showing that initial dehydration melting of an amphibole- and/or biotite-bearing tonalite and granodiorite source produces ferroan granitic melts within a compositional range of A-type granites (e.g., Skjerlie and Johnston, 1993; Patiño Douce, 1997). The tonalite-granodiorite source model also gained support from the general observation that the emplacement of A-type suites commonly postdate the formation of other granite types (e.g., I- and S-type granite), either in an intraplate and rift setting or at a late stage in the orogenic cycle (Creaser et al., 1991; Eby, 1992).

It was recognized early on that the peak temperatures of A-type granite melts are high during their emplacement (~900 ºC) (Clemens et al., 1986; Creaser et al., 1991; Skjerlie and Johnston, 1993; Patiño Douce, 1997) and attainment of such high temperatures requires intrusion and crystallization of large volumes of mantle-derived mafic melts in the continental crust. Contemporaneity of A-type granite suites and associated mafic igneous rocks has been pointed out in numerous studies (e.g., Bonin and Giret, 1990; Eby et al., 1992; Turner et al., 1992; Rämö and Haapala, 1995; Smith et al., 1999a,b). A-type granites generally occur in the tectonic regimes in which the crust is undergoing extension (e.g., continental rifts, back arc rifts, postcollisional settings etc.), triggering decompressional melting of the mantle and allowing mantle-derived melts to rise into the Earth’s crust. Several studies also recognized that hot mafic magmas may not only transfer heat for crustal melting to generate A-type granite melts but also produce A-type granitic melts via differentiation with or without assimilation of partial melts from the crust through which they ascend (e.g., Capaldi et al., 1987; Kleeman and Twist, 1989; Rämö, 1991; Eby et al., 1992; Tommasini et al., 1994; Chazot and Bertrand, 1995; Smith et al., 1999a,b). More recently, several studies have shown that interaction between juvenile material and older crust is indeed significant in many A-type suites (Schmitt et al., 2000; Kemp et al., 2005, 2009; Frost and Frost, 2011). Frost and Frost (2011), for example, propose that peralkaline ferroan A-type granites form by differentiation of transitional or alkali basalt, potentially with some crustal assimilation, and metaluminous ferroan granite may form by differentiation of tholeiitic basalt with or without crustal assimilation and partial melting of quartzo-feldspathic crust at low pressure. Ferroan granites also become increasingly more peraluminous by involvement of partial melts of quartzo-feldspathic crust (Frost and Frost, 2011). Other researchers suggest that some A-type granites may have been generated by direct fractional crystallization of mafic parental magmas, without involving a crustal component (e.g., Shellnutt et al., 2011; Shellnutt and Dostal, 2020). Some researchers have proposed that an A-type granite melt composition may also arise by partial melting of a lower crustal source preconditioned by fluids from degassing mantle in a lithospheric delamination regime (e.g., Martin, 2006, 2012; Bastos Neto et al., 2010) or by partial melting of underplated mafic rocks, triggered by the upwelling asthenosphere (e.g., Frost et al., 1999; Shellnutt and Zhou, 2007; Su et al., 2017; Zong et al., 2020).

In summary, the studies on the petrogenesis of A-type granites have proposed three main modes of origin: 1) partial melting of meta-igneous crustal rocks, such as tonalite or granodiorite, 2) extensive fractionation of mantle-derived magmas, involving removal of large amounts of silica-poor mineral assemblages, and 3) a combination of the first two models, in which differentiating mantle-derived magmas assimilate crustal rocks. These processes are not limited to any specific tectonic setting as, for example, crustal melting can consequence from several tectonic processes and, in principle, any extensional setting that opens a window to the mantle can be regarded as favorable for mantle-derived magmatism and coeval emplacement of A-type granites.

2.4 REE-HFSE mineralization in A-type granites

A-type granite-hosted REE-HFSE deposits are typically associated with peralkaline granites and related rocks corresponding to the A1-type of Eby (1992) (e.g., Kovalenko et al., 1995; Dostal and Shellnut, 2016; Gladkochub et al., 2017; Vasyukova and Williams-Jones, 2020). The peralkaline granite plutons that host REE-HFSE deposits are typically relatively small in size, ranging from 3 to 25 km in diameter. They occur in continental rift settings where their locations are commonly controlled by fault zones related to regional extension (Kovalenko et al., 1995; Siegel et al., 2017a). The REE-HFSE mineralization usually occurs in highly evolved peralkaline granite stocks or pegmatites and/or aplite sheets and associated metasomatic rocks hosted within or near roofs of plutons. The reported mineral resources range from tens to hundreds of millions of tonnes with the typical grades being ca. 0.3 to 1.5 wt% for TREE, 0.3

to 5 wt% for Zr and <1 wt% for other HFSE (e.g., Nb, Ta, Hf, Th, U) (Kovalenko et al., 1995; Yang et al., 2014; Gysi et al., 2016).

Chemically, evolved REE-HFSE-rich peralkaline felsic rocks have elevated Fe contents and high Fe/Mg and Ga/Al, and compared to most other granites, show relative enrichment in the heavy REE (e.g., Dostal, 2016), depletion in Eu, and enrichment in halogens, especially F, and U and Th. The host igneous rocks and immediate country rocks of mineralization frequently display hydrothermal alteration halos (e.g., K-Na-, Na-Ca-, or Ca-F-metasomatism and REE and HFSE dispersion) due to escape of late hydrothermal fluids from the magma parental to mineralization (e.g., Schmitt et al. 2002; Salvi and Williams-Jones 2005; Williams-Jones et al., 2012; Dostal et al., 2014; Gysi et al., 2016; Morgerstern et al., 2018). Typical rock-forming minerals in the host REE-HFSE-rich peralkaline granites include quartz, alkali feldspar and Na-rich clinopyroxene and/or amphibole, which in some cases are perplexed by pervasive hydrothermal fluid alteration. Rare earth elements and HFSE may be present in a wide range of accessory mineral species, which contain REE-HFSE as an essential component in their crystal structure. The assemblages of REE- and HFSE-bearing minerals are typically widely variable within individual deposits and between deposits, as some of them contain miaskitic assemblages (e.g., Kovalenko et al., 1995; Sun et al., 2013), in which the main REE-HFSE-bearing minerals are zircon, allanite, titanite and oxides (e.g., pyrochlore), whereas others have agpaitic assemblages (exotic Na-Ca-Ti-Zr silicates, e.g., eudialyte, elpidite; e.g., Kynicky et al., 2011) and some record a transitional mineral assemblages with early formed miaskitic to late agpaitic phases (e.g., Estrade et al., 2014). Most peralkaline REE-HFSE-mineralized granites are dominated by miaskitic REE-HFSE mineral assemblages, and only a very small group of these rocks contain mainly agpaitic minerals (Marks et al., 2011; Vasyukova and Williams-Jones, 2020).

Enrichment of REE-HFSE in peralkaline A-type granites to potentially economic levels is commonly considered to require a combination of various source and magma chamber processes and late magmatic hydrothermal remobilization (Schmitt et al., 2002; Salvi and Williams-Jones, 2005; Sun et al., 2013; Kynicky et al., 2011; Kempe et al., 2015; Dostal, 2016; Vasyukova and Williams-Jones, 2020). Because of their high contents of incompatible trace elements, alkalis and halogens, the parental melts to peralkaline felsic rocks fertile to host REE-HFSE deposits are typically thought to origin by a very low degree of melting of a mantle plume or metasomatically enriched mantle below a continental crust (continental rift) (Kemp et al., 2005; Kovalenko et al., 2007; Kogarko et al., 2010; Wu et al., 2010; Frost and Frost, 2013;

Gerdes et al., 2017; Vasyukova and Williams-Jones, 2020). The source rocks in the mantle are likely amphibole- or phlogopite-bearing as indicated by the high contents of halogens in REE-HFSE-rich peralkaline rocks (Chakhmouradin and Zaitsev, 2012). It is probable that peralkaline felsic rocks are formed largely by differentiation of transitional or alkali basalt (e.g., Barberi et al.,1975; Brotzu et al.,1983; Eby et al., 1992; Ronga et al., 2010; Shellnutt et al., 2011; Shao et al., 2015; Jeffery and Gertisser, 2018). However, it has also been proposed that crustal contamination of silica-undersaturated nephelinitic parental melts may result in peralkaline granite (e.g., Goodenough et al., 2000; Marks et al., 2003; Vasyukova and Williams Jones, 2020).

High-contents of REE-HFSE in peralkaline felsic rocks are, at least in part, due to protracted crystal fractionation. Mantle-derived nephelinitic to basaltic melts can evolve to compositions similar to those of peralkaline felsic rocks with appreciable REE-HFSE concentrations by protracted fractional crystallization involving feldspar-dominated fractionation at a relatively low pressure (e.g., Barberi et al., 1975; Eby et al., 1992; Zozulya et al., 2009; Ronga et al., 2010; Shellnut et al., 2011; Dostal and Shellnut, 2016; Jeffery and Gertisser, 2018). Fractionation plays a crucial role in REE-HFSE concentration because REE and HFSE behave as highly incompatible elements with very low solid/melt partition coefficients (<1) for most part of the fractionation process (e.g., Linnen et al., 2014). They do not enter easily into the crystal structure of common rock-forming minerals (e.g., feldspar, olivine, pyroxenes) and therefore become preferentially enriched in the residual melt. Chemical changes in the magma composition during fractional crystallization contribute to the accommodation of REE-HFSE in the melt. Fractionation of a basaltic magma that proceeds at oxygen fugacities well below the fayalite-magnetite-quartz oxygen buffer (Frost and Lindsley, 1992) suppresses the crystallization of magnetite and results in early removal of Si-poor and Mg-, Ca- and Fe-rich silicates (e.g., Mg-rich olivine and pyroxenes) and an increase in the Al_2O_3 content of the melt until calcic plagioclase starts to crystallize as one of the liquidus phases. This will lead to an increase in SiO_2, Na_2O and K_2O and Fe/Mg and a decrease in CaO and Al_2O_3 in the residual melt and also in a depletion of Eu (Eu^{2+} substitutes for Ca^{2+} in plagioclase in reduced conditions) that is a characteristic feature of REE-HFSE-rich peralkaline granites. Removal of abundant calcic plagioclase increases peralkalinity in the residual melt, enabling incorporation of REE-HFSE in the magma by increasing the saturation threshold of typical accessory REE-HFSE-bearing minerals (e.g., monazite, allanite, zircon) until the last stages of crystallization. The reason for this behavior of REE-HFSE is that the excess alkali ions (Na^+ and K^+) act as network modifiers when their proportions exceed that of Al to substituting Si in the silicon tetrahedron (Vasyukova and Williams-Jones, 2020). This reduces the degree of melt polymerization by increasing the ratio of non-bridging oxygen atoms to network-forming tetrahedrally coordinated cations, such as Si and Al, which reduces the available SiO_2 tetrahedra to form accessory REE-HFSE minerals, stabilizing accommodation of REE-HFSE in the melt (Watson, 1979; Montel, 1986; Peiffert et al., 1996; Linnen and Keppler, 1997; Aseri et al., 2015). The large size of the Na^+ and K^+ ions (ionic radii of 1.18 and 1.52 Å, respectively) also prevents much smaller ions (e.g., Zr^{4+} 0.84 Å) from bonding with the available SiO_2 tetrahedra and to form accessory minerals (e.g., zircon; Watson, 1979; Montel, 1993). Moreover, crystallization of clinopyroxene, olivine, and calcic plagioclase increases the activity of volatile components (e.g., OH^-, F^-, Cl^-, SO_4^{2-}, CO_3^{2-}), which are easily dissolved in alkali-rich silicate melts (e.g., Collins et al., 1982; Keppler, 1993; Carroll and Webster, 1994; Agangi et al., 2010). Volatile ions may further raise the saturation threshold of REE-HFSE as they also potentially act as network modifiers (e.g., Keppler, 1993; Farges, 1996; Giordano et al., 2004; Zimova and Webb, 2007).

The role of crustal contamination in the genesis of REE-HFSE-rich peralkaline felsic rocks is still debatable. Peralkaline granitic magma cannot be readily derived by partial melting of common crustal source rocks, which are typically peraluminous (Bonin, 2007). However, oxygen and Nd isotopic evidence suggests that most REE-HFSE-rich peralkaline granites record contribution of crustal material to their magmas (e.g., Kerr and Fryer, 1993; Jahn et al., 2001; Schmitt et al., 2000; Trumbull et al., 2004; Kovalenko et al., 2007; Siegel et al., 2018). Assimilation is considered a key process in driving mantle-derived parental melts to evolve to granitic compositions by introducing additional SiO_2. In addition, in the case of a silica-undersaturated parental melt, assimilation is essential in causing the melt to crystallize feldspars instead of their silica-undersaturated counterparts (e.g., nepheline and leucite) (Vasyukova and Williams-Jones, 2020). As the continental crust is enriched in REE-HFSE relative to the mantle (Rudnick and Gao, 2003, 2014), it is possible that crustal assimilation is an important mechanism to add REE-HFSE to the magma, which could also satisfactorily explain the consistently low initial ε_{Nd} values of many REE-HFSE-rich A-type granites (e.g., Goodenough et al., 2000; Schmitt et al., 2000; Marks et al., 2003; Trumbull et al., 2004; Jahn et al., 2009; Siegel et al., 2017a).

Several processes related to crystallization of REE-HFSE-rich peralkaline granitic magmas can also contribute to hyper-enrichment in REE-HFSE. Peralkaline granites crystallize massive amounts (~50 to 70 wt%) of alkali feldspar typically rich in K (e.g., Schmitt et al., 2002; Siegel et al., 2018; Vasyukova and Williams-Jones, 2020), resulting in a progressive increase in the Na content and Na/K ratio of the residual magma. This ensures a continuous increase in the concentration of REE-HFSE in the magma by helping it to become more depolymerized. The increase in the Na activity coupled with an elevated activity of volatiles (e.g., OH^-, F^-) and high Fe/Mg is likely to promote crystallization of Na- and Fe-rich sodic amphibole (e.g., arfvedsonite), which potentially has a large impact on the REE-HFSE enrichment in the residual melt because amphibole crystallization consumes OH^- and postpones aqueous phase exsolution (Vasyukova and Williams-Jones, 2020). In addition, amphibole accommodates more Fe relative to Na, which may further increase the Na concentration in the residual melt and the solubility of REE-HFSE-bearing accessory minerals (Vasyukova and Williams-Jones, 2020). This contributes to continued enrichment of REE-HFSE as preservation of Na^+ and halogens in the melt increases the solubility of REE-HFSE, preventing their loss to an exsolving magmatic fluid phase. The increase in the Na concentration results in a several orders of magnitude increase in the solubility of REE-HFSE minerals (e.g., zircon), allowing the incorporation of REE-HFSE in the residual magma at degrees required for formation of a potentially economic deposit. The overall effect is that the magma is eventually transformed to crystallize agpaitic mineral assemblages instead of simple miaskitic ones.

In addition to fractional crystallization, a process that has recently been shown to play an important role in the development of potentially mineable peralkaline granite-related REE-HFSE ore bodies is silicate-fluoride liquid immiscibility (Vasyukova and Williams-Jones, 2014, 2020). With decreasing temperature, silicate melts are shown to exsolve a fluoride liquid, from which fluorite crystallizes (Yang and van Hinsberg, 2019). If a granitic magma is split into a silicate melt and a fluoride melt, REEs will partition preferentially to the latter (Vasyukova and Williams-Jones, 2014). In order of this process to result in additional REE enrichment, the immiscibility should occur early and the fluoride melt crystallization late in the crystallization history of a pluton. Although the factors responsible for silicate-fluoride immiscibility are not yet well understood, it is proposed that this phenomenon can only develop in a putative parental magma which undergoes a rapid change from calcic plagioclase to alkali feldspar crystallization prior to the final consumption of Ca, possibly due to a rapid ascent of the magma (Vasyukova and Williams-Jones, 2020). This may induce a sudden increase in the Ca activity in the residual melt and saturation of an immiscible Ca-fluoride melt, but only if the F activity in the melt is sufficiently high (Vasyukova and Williams-Jones, 2020). Once exsolved, the fluoride liquid interacts with the adjacent silicate liquid, strongly depleting it in Ca and REEs and enriching it in HFSE, such as Zr and Nb, which prefer the conjugate residual silicate melt (Vasyukova and Williams-Jones, 2014). As the fluoride melt is only able to crystallize late in the crystallization history of granitic magmas, this process can result in sequestering REE in the granite magma to a late magmatic stage, causing hyper-enrichment of REE in the volatile-rich last residue.

Formation of a peralkaline granite-hosted REE-HFSE deposit also greatly depends on the saturation of the magma with an aqueous-carbonic fluid in very last stages of magmatic evolution. In several REE-HFSE-rich peralkaline granites, exsolved magmatic fluids have pervasively altered and mineralogically re-equilibrated granites and associated pegmatites-aplites at relatively low temperatures (e.g., Kempe et al., 2015; McDonald et al., 2015; Siegel et al., 2018). In some cases, related remobilization of REE-HFSE has led to replacement of the primary (magmatic) REE-HFSE-bearing minerals with secondary low-temperature assemblages, increasing considerably the resources of the deposit (e.g., Gysi et al., 2016). The ideal scenario is one in which the initial fluid is extremely reduced and CH_4-dominated and evolves to CO_2 saturation. To be an effective mobilizer of REE and HFSE, the exsolved fluid also needs to evolve to a very low pH, which allows it to decompose primary REE-HFSE-bearing phases and release their elements in the fluid. However, extensive decomposition of primary minerals concerns only the agpaitic mineral assemblages as the miaskitic ones are much more refractory to mobilization of REE and HFSE. In most favorable cases, redeposition of REE takes place as REE fluorocarbonates (e.g., Kynicky et al., 2011; Estrade et al., 2014; Kempe et al., 2015), from which REE can be efficiently recovered by established hydrometallurgical methods (e.g., Verbaan et al., 2015; Weng et al., 2015; Demol et al., 2019; Peiravi et al., 2021).

3 Regional geology of the Otanmäki area

The Otanmäki area is located within the southwestern margin of the Archean Karelia craton, central Fennoscandian Shield, approximately 40 km to the east from the abrupt tectonic boundary, which the craton has against the Svecofennian domain consisting almost solely of Paleoproterozoic assemblages. The location is 30–50 km to the west from the Paleoproterozoic cover sequences (the Karelian formations) preserved in the Kainuu belt (Figs. 1a and b). The long geological history of the area involves at least four major tectonic stages: 1) Archean orogeny at ca. 2.8–2.6 Ga, 2) a period of cratonization, followed by long-lived stable cratonic sedimentation, interrupted by minor mafic dike swarms produced by extensional tectonic events between ca. 2.45 and 2.0 Ga, 3) subsequent continental break-up of the Karelia craton between ca. 2.0 and 1.95 Ga, and 4) tectonic reworking at ca. 1.92–1.77 Ga in connection with a complex set of orogenic events assigned to the Svecofennian orogeny (Laajoki, 2005; Vuollo and Huhma, 2005; Lahtinen et al., 2010, 2015; Huhma et al., 2012; Hölttä et al., 2012a,b; Davey et al., 2020). The present structural configuration of the area largely reflects stage 4, which involved several episodes of compressional folding and thrusting related to the burial of the southwestern margin of the Karelia craton under a massive overthrust complex transported from the west-southwest, and a subsequent phase of strike-slip tectonics (e.g., Kontinen et al., 1992, Lahtinen et al., 2015; Hölttä et al., 2019a).

The Archean bedrock of the area around the Otanmäki area is composed mostly of TTG migmatite-gneiss complexes overlain by remnants of Paleoproterozoic cover sequences preserved in the Kainuu belt (Laajoki, 2005, Hölttä et al., 2012a,b, 2019b). The Archean complexes of the area belong to the Manamansalo and Iisalmi complexes composed mostly of TTG gneisses and the Kajaani complex composed of gneissic supracrustal and TTG rocks (ca. 2.6–3.4 Ga) (Hölttä et al., 2012a, 2019b). The supracrustal sequence of the Kainuu belt is traditionally divided into three main unconformity-separated units, Sumi-Sariola, Jatuli, and Kaleva (Laajoki, 2005; Lahtinen et al., 2015). The metasupracrustal rocks of the Kainuu belt flanking the Otanmäki area in the east belong to the Jatulian (2.1–2.3 Ga) and Kalevian (1.9–2.1 Ga) formations. The Jatuli consists of cratonic-epicratonic deposits, and the Kaleva includes both rift-phase marine basin deposits (Lower Kaleva) and deep-water oceanic basin sedimentary rocks (Upper Kaleva). The Upper Kaleva sediments differ from the Jatulian and Lower Kaleva sedimentary rocks, which were derived dominantly from Archean sources, in having a major Paleoproterozoic (1.92–2.0 Ga) detrital zircon population (Lahtinen et al., 2010). The Upper Kaleva wackes enclose fragments of mantle and oceanic crust, with the largest of them being the 1.95 Ga Jormua ophiolite complex (Kontinen, 1987; Peltonen and Kontinen, 2004). The Upper Kaleva metasediments and enclosed ophiolites have been interpreted as an allochthonous complex, which was thrust on the Karelia craton margin at ca. 1.9 Ga (Peltonen and Kontinen, 2004).

The area around the Otanmäki area also records several Paleoproterozoic magmatic episodes (Fig. 1b) as it contains a 2.44 Ga layered intrusion (Junttilanniemi intrusion; Halkoaho and Niskanen, 2013; Huhma et al., 2018), 2.3 Ga mafic-ultramafic intrusions (Kapustakangas suite; Huhma et al., 2018), 2.2–2.1 Ga mafic dikes and volcanic rocks (Huhma et al., 2018), 2.06 Ga

Fe-Ti-V magnetite-ilmenite ore-bearing mafic intrusions (Otanmäki intrusive complex; Huhma et al., 2018; Mäkisalo, 2019), ca. 2.05 Ga A1-type intermediate to felsic intrusions (Otanmäki suite; Papers I–II), and at least two major episodes of late Svecofennian granitic magmatism dated at ca. 1.86 and 1.80 Ga (Kontinen et al., 2013b).

The Svecofennian metamorphic conditions in the Otanmäki area are estimated to have attained peak temperatures of ~550–600 ºC and a pressure of about ~4.0 kbar (Paper I), corresponding to the main Svecofennian metamorphic event (e.g., Hölttä and Heilimo, 2017). The most plausible explanation to the observed Svecofennian heating is tectonic burial of the presently exposed rocks to depths of 15–20 km in association with the 1.8–1.9 Ga folding and thrusting events, although igneous underplating could also have been an additional factor (Kontinen et al., 1992, 2013b,c; Tuisku, 1997; Korsman et al., 1999; Pajunen and Poutiainen, 1999; Kontinen, 2002; Kontinen and Paavola, 2006; Lahtinen et al., 2010, 2015). The peak-T conditions are considered to have occurred between ca. 1.87 Ga and 1.85 Ga, although the heating was protracted, with the temperatures falling below ~500 ºC only after 1.80 Ga (Kontinen et al., 1992; Vaasjoki et al., 2001; Hölttä et al., 2019a).

4 Previous studies of the Otanmäki suite and related REE-HFSE mineralization

The granitic rocks at Otanmäki drew attention already during the pioneering regional mapping work of Wilkman (1929, 1931), who was the first to describe occurrences of gneissic, reddish hornblende granite containing hornblende-rich bands and inclusions in an area south of Lake Oulujärvi. These unusual granites were first studied in detail when the Otanmäki area became targeted by systematic exploration and geological mapping after the discovery of the Otanmäki magnetite-ilmenite ore deposit in 1938 (Paarma, 1954).

Pääkkönen (1956) provided the first detailed description of the geology of the Otanmäki area, including a description of the special granite variety in the vicinity of the gabbro bodies hosting the Fe-Ti-V oxide ore bodies. This granite type was characterized by a gneissic texture, the presence of alkali amphibole and an unusually high content of total alkalies and FeO_{tot} compared to other granites known from Finland at the time (Pääkkönen, 1956). Marmo et al. (1966) were the first to make a more detailed study of the 'alkaline granite' at Otanmäki and showed that it had a peralkaline bulk composition and contains alkali amphibole and occasional alkali pyroxene (aegirine). The alkaline granite in the vicinity of the Otanmäki Fe-Ti-V mine was also mapped by geologists of the Rautaruukki Oy mining company (e.g., Lindholm and

Anttonen, 1980), who recognized that the alkaline granite is closely associated with the quartz-feldspar gneiss unit that was later discovered to host REE-HFSE enrichments (see below).

The geological setting of the Otanmäki alkaline granite remained obscure until it was gradually mapped by GTK geologists in various bedrock mapping or other research projects (e.g., Peltonen et al., 1996; Havola, 1997; Kontinen and Paavola, 2006; Kontinen et al., 2013a). The mapping results indicated that the alkaline granite is not only limited to the vicinity of the Otanmäki Fe-Ti-V mine but instead occupies a E- to W-trending, 1- to 8-km-wide and 60-km-long boomerang-shaped area, which extends from Otanmäki (west) via Mainua, Lehtovaara, Kontinjärvi, Soidinvaara to Kuluntalahti (east) (Fig. 1b). Moreover, these studies provided the first clues to the geological setting of the alkaline granite. It was observed to be bordered by Archean gneiss complexes and metasedimentary units of the Karelian formations and extensively intruded by younger Svecofennian granites, and its proximity to the Jormua ophiolite complex was noted.

Age determinations of the alkaline granite were made in several stages in the GTK. Zircon grains separated from a granite sample A100 taken by Marmo et al. (1966) from the Otanmäki area (Fig. 1b; Table 1) were analyzed by O. Kouvo in the 1960s using U-Pb thermal ionization mass spectrometry (TIMS), giving an age of ca. 2.05 Ga (Marmo et al., 1966). The age was later refined by O. Kouvo using more zircon fractions of the same sample, which jointly with the older data gave a slightly younger U-Pb age estimate of ca. 2.02 Ga (Hytönen and Hautala, 1985). In addition, a zircon U-Pb age of 2.03 Ga, which remained unpublished, was produced by O. Kouvo in the 1970s, using zircon fractions from a granite sample (A381) taken by M. Asa and K. Meriläinen from the Kuluntalahti area (Fig. 1b; Table 1). Significantly younger U-Pb ages of ca. 1.96 Ga were obtained in the 1990s based on multigrain TIMS dating of zircon grains separated from the two granite samples (A1149 Otanmäki and A989 Soidinvaara; Fig. 1b, Table 1) picked for analysis by M. Vaasjoki and M. Havola. The ca. 1.96 Ga ages remained unpublished but were particularly interesting as they placed the Otanmäki alkaline granite temporally near the Jormua ophiolite complex (ca. 1.95 Ga), as was pointed out by Peltonen et al. (1996). Peltonen et al. (1998) suggested that the emplacement of the Otanmäki alkaline granite was linked to the initial rupture of the craton and coeval OIB-like basaltic magmatism, which preceded the seafloor spreading and oceanic crust forming magmatism recorded by the 1.95 Ga ophiolites.

Table 1. Summary of U-Pb zircon dating results.

Sample	Location	Northing	Easting	Rock type	Age/U-Pb TIMS Produced previously	Age/U-Pb CA-TIMS Paper I	Age/U-Pb LA-ICP-MS Paper I	Age/U-Pb SIMS Paper III
Western part of the Otanmäki-Kuluntalahti nappe								
#1 A100	Honkamäki, Kajaani	7115100	3497400	Peralkaline granite, coarse-grained	c. 2050[1] *2020 ± 5[2]		2041 ± 5	
#2 A1149	Otanmäki, Kajaani	7113778	3505350	Peralkaline granite, coarse-grained	*1958 ± 29[3]		2049 ± 10	
#3 160271	Pieni-Katajakangas, Kajaani	7114244	3501926	Peraluminous granite, fine-grained			2055 ± 8	
#4 160270	Pieni-Katajakangas, Kajaani	7114166	3501918	Peraluminous granite, fine-grained			2060 ± 29	
#5 KA-R9-6.25-6.70m	Luodesuo, Kajaani	7115342	3502186	Kontioaho REE-HFSE mineralization				2036 ± 4
Eastern part of the Otanmäki-Kuluntalahti nappe								
#6 A1896	Kontinjärvi, Sotkamo	7114975	3541384	Peraluminous granite, coarse-grained		2050 ± 2	2053 ± 11	
#7 A1832	Kontinjoki, Sotkamo	7117515	3542542	Peraluminous granite, coarse-grained		2046 ± 3		
#8 A989	Soidinvaara, Sotkamo	7117140	3543380	Peraluminous granite, fine-grained	*1965 ± 13[3]			
#9 A381	Kuluntalahti, Kajaani	7129110?	3544400?	Peraluminous granite, fine-grained	*ca. 2030[3]			
Lehtovaara lens								
#10 A1833	Käärmekalliot, Kajaani	7108790	3532953	Peraluminous granite, coarse-grained		2047 ± 3		
#11 A1834	Niskalankalliot, Kajaani	7108099	3532151	Peraluminous granite, fine-grained		2047 ± 5		

*Age reflects metamorphic effects; see Paper I for details. Coordinates in KKJ-3/Finland Uniform Coordinate System. For sample locations on map, see Fig. 1b.

References: [1] Marmo et al. (1966), [2] Hytönen and Hautala (1985), [3] GTK (unpublished)

Kontinen et al. (2013a) recognized that the “alkaline granite” consists of a distinct chemical suite of peralkaline and peraluminous granites (the Otanmäki suite) plotting in the fields of within-plate granites and A1-type granites with an OIB-like trace element affinity on granite discrimination diagrams, thus confirming the similarity with the Red Sea 20–30 Ma granites as proposed by Peltonen et al. (1996). They reported new U-Pb zircon age data, which indicated an age of ca. 2050 Ma for the Otanmäki suite granitic magmatism, and their emplacement up to 100 Ma earlier than was proposed previously by Peltonen et al. (1996, 1998). Neodymium isotope data from representative granite samples yielded mostly mantle-like ε_{Nd}(2050 Ma) values ranging from −3.1 to +0.7. Based on these data, Kontinen et al. (2013a) proposed that the origin of the Otanmäki suite granitic rocks probably involved an OIB-like mantle source and variable contamination during magma ascent through lithosphere and extended/faulted continental crust.

The REE-HFSE enrichments in the Otanmäki area (Fig. 1b) were first studied by the GTK and Rautaruukki Oy during the first exploration stage between 1981 and 1985. They were briefly re-evaluated during GTK’s high-tech metal project between 2009 and 2011 (Sarapää et al., 2013, 2015) and further explored by Tasman Metals Ltd between 2010 and 2013, though all their geological and mineralogical descriptions preceding this work are restricted to brief notes in old exploration reports (Hugg, 1985a,b; Hugg and Heiskanen, 1986). Rautaruukki Oy reported that the two REE-HFSE-mineralized zones occur as shallow-dipping, concordant layers with sharp contacts to their quartz-feldspar gneiss wall rocks, reaching several hundreds of meters in strike length and at least 150−200 m in depth. The thickness of these layers was determined to be 0.2−1.4 m at Katajakangas and 7−12 m at Kontioaho, and the following REE-HFSE-bearing minerals were identified: allanite, columbite, fergusonite, xenotime, titanite, and zircon (Hugg, 1985a,b). The quartz-feldspar gneiss wall rock was interpreted to represent Proterozoic metapsammite, which was metasomatized by alkaline fluids (Hugg and Heiskanen, 1986). A metasomatic origin was also suggested for both the REE-HFSE-mineralized rock units and nearby ‘alkaline granite’, but without any proposition for the source of the hypothesized metasomatizing alkaline REE-HFSE-rich fluids (Hugg and Heiskanen, 1986). Later Kontinen et al. (2013a) proposed that instead of metasomatic rocks, the ‘alkaline granite’ is a peralkaline A1-type granite and the quartz-feldspar gneiss wall rock was possibly a co-genetic felsic volcanic or subvolcanic rock.

5 Methods and sampling

Field work, including sampling for Papers I–III, was carried out during the time period of 2016–2018. For Papers I and III, material was also collected by A. Kontinen in 2005–2006. Totally, the dataset contains 7500 field observations. In addition, 31 diamond drill cores from the Otanmäki suite igneous rocks and REE-HFSE-mineralized rock bodies were studied at the Finnish National Drill Core Archive located at Loppi, southern Finland, and archived data on a total of 103 diamond drill cores were also utilized (for list of diamond drill cores and their locations see Appendix).

A polarized light microscope with transmitted and reflected light capabilities was used at the University of Oulu to study approximately 750 polished thin sections. Petrographic classification of the Otanmäki suite igneous rocks was based on modal compositions obtained by point counting a minimum of 1000 points and the IUGS QAP scheme (Streckeisen, 1967). Chemical compositions of REE-HFSE-bearing minerals were determined by wavelength dispersive X-ray spectroscopy (WDS) at the Helmholtz-Institute Freiberg (HIF) with a JEOL JXA-8530F electron probe microanalyzer (EPMA) and JEOL JXA-8200 EPMA at the Centre of Material Analysis, University of Oulu. For the description of the quality control procedures and analytical conditions, see Appendices in Papers I and III. In addition, at the HIF, a FEI Quanta 650 F field emission scanning electron microscope (FE-SEM) with two Bruker Quantax X-Flash 5030 energy-dispersive X-ray detectors (EDS) was employed for semi-automated scanning of polished thin sections and FEI's Mineral Liberation Analyzer (MLA) software suite 3.1.4 for data acquisition, processing and evaluation.

Multi-grain zircon U-Pb analyses for Paper I were performed using a VG Sector 54 thermal ionization mass spectrometer and isotope dilution (ID-TIMS). The method of the U-Pb TIMS analysis followed the procedures described in Huhma et al. (2018) and the analytical quality was on par with that in the given paper. In addition, in order to better constrain the effects of metamorphism on the zircon ages, the chemical abrasion (CA) TIMS method of Mattinson (2005) was utilized. Single grain U-Pb isotope analyses for Paper I were also made on zircon at the Finnish Isotope Geosciences Laboratory hosted by GTK in Espoo using a Nu Plasma high resolution (HR) multicollector (MC) ICP-MS instrument or a Nu Plasma AttoM single collector (SC) ICP-MS instrument connected to a Photon Machine Excite laser ablation (LA) system (LA-MC-ICP-MS and LA-SC-ICP-MS methods, respectively). The methods and related analytical quality control protocols are described in Huhma et al. (2018). Zircon single-grain U-Pb age determinations for Paper III were conducted at the Nordsim laboratory, Museum of Natural History of Sweden, Stockholm, using a Cameca II IMS 1270 secondary ion mass spectrometer (SIMS) and an analytical procedure similar to that described by Jeon and Whitehouse (2015) and references therein. The age calculations were performed and concordia diagrams for Papers I and III made using the Isoplot/Ex 4.15 program (Ludwig, 2008).

Whole-rock Sm-Nd isotope analyses for Papers II and III were performed in three different stages. The chemical compositions of the first sample batch were measured at the GTK using ID-TIMS as described by Huhma et al. (2018). The second batch was analyzed at ALS Scandinavia AB using inductively coupled plasma-sector field mass spectrometry (ICP-SFMS), following its standard procedure including digestion of samples by alkali fusion, dissolution in a $C_4H_6O_6$-HNO_3 mixture and separation of Sm and Nd by ion exchange chromatography. The third batch was analyzed at the Finnish Isotope Geoscience Laboratory at GTK using high-resolution multicollector ICP-MS after sample digestion in a HF-HNO_3 mixture and separation of Sm and Nd by ion exchange chromatography. Further details on the ICP-SFMS and HR-MC-ICP-MS analytical procedures and accuracy are given in Electronic Appendices of Papers II and III.

Thermodynamic and geochemical modeling in Paper II was carried out using the Magma Chamber Simulator (MCS) code (Bohrson et al., 2014, 2020; https://mcs.geol.ucsb.edu/). MCS enables the user to study assimilation and fractional crystallization (AFC) processes in a multicomponent-multiphase silicate magma-wall rock system using the MELTS family of models (Ghiorso and Sack, 1995; http://melts.ofm-research.org/) as its engine to derive phase equilibria. Based on thermodynamic data and constraints, MELTS employs variations in temperature, pressure, and volume to replicate the thermodynamic and chemical evolution of a magmatic system undergoing (equilibrium or fractional) crystallization or partial melting. Rhyolite-MELTS version 1.1.0 was used as the thermodynamic engine of MCS for modeling the Otanmäki suite igneous rocks because it has been optimized for silicic systems that saturate in quartz and crystallize to the two-feldspar-, quartz-, fluid-saturated ternary minimum within the SiO_2-TiO_2-Al_2O_3-Fe_2O3-Cr_2O_3-FeO-MnO-MgO-NiO-CoO-CaO-Na_2O-K_2O-P_2O_5-H_2O compositional space (Gualda et al., 2012; Ghiorso and Gualda, 2015).

6 Review of the original publications

6.1 Paper I

Paper I presents field and petrographic observations, geothermobarometry and whole-rock geochemical and U-Pb zircon and titanite isotope data from the previously only superficially studied Otanmäki suite of ‘alkaline granites’. The aim of this paper is to characterize the rock types of the Otanmäki suite and their spatial distribution and to conduct new multigrain TIMS and single-grain LA-MC-ICP-MS zircon U-Pb dating in order to constrain the nature and age of the magmatism and make conclusions on the geotectonic environment of the Otanmäki suite.

Although the areal extent of the Otanmäki suite has been roughly known since 2013 (see Kontinen et al., 2013a), the distribution of the main rock types within the suite is properly presented for the first time in Paper I. The study reveals that the Otanmäki suite consists of a spectrum of ferroan peralkaline to peraluminous granites and previously unrecognized intermediate igneous rocks including syenite and monzodiorite-monzonite (Figs. 2 and 3). The bulk of these igneous rocks is located in an E-W-trending, approximately 60-km-long and 1– to 10-km-wide belt (approximately 250 km^2 in area) (Fig. 1 b). Being obviously thrust fault-bound on both sides, it is named the Otanmäki-Kuluntalahti nappe, representing the main nappe unit. The nappe is bordered in the east by the Karelian formations of the Kainuu belt and in north and the west, by Archean gneiss complexes. The narrowest central part of the main nappe is confined between Archean gneisses in the north and a narrow strip of Karelian intercalated metabasalts, gabbroic sills and metasedimentary rocks in the south. In addition, approximately 0.5 km south of the central segment of the main nappe, A1-type rocks of the Otanmäki suite are observed to intrude Proterozoic quartzites in a 3x8-km-sized, fault-bound and lens-shaped body called the Lehtovaara lens (Fig. 1b).

The westernmost part of the main nappe consists mainly of peralkaline granite and, to a lesser extent, syenite and monzodiorite-monzonite, which are characterized by coarse-grained plutonic textures and presence of mafic enclaves. These rock types are also mingled with blocks of K-feldspar-phyric, fine-grained, subvolcanic-type peraluminous granite, with the largest of them enclosing up to 200-m-thick and 5-km-long slivers of Lower Kaleva-type metapsammite-pelite schists (Fig. 1b). LA-MC-ICP-MS analyses of zircon fractions from two samples of the peralkaline granite (A1149 and A100) yield ages of 2049 ± 10 Ma and 2041 ± 5 Ma (Fig. 1b; Table 1), which are interpreted as probable magmatic ages.

Two samples of the peraluminous granite, 160271 and 160270, dated using in-situ LA-SC-ICP-MS analysis give U-Pb zircon ages of 2055 ± 8 Ma and 2060 ± 29 Ma, respectively, also considered ages of magmatic crystallization (Fig. 1b; Table 1).

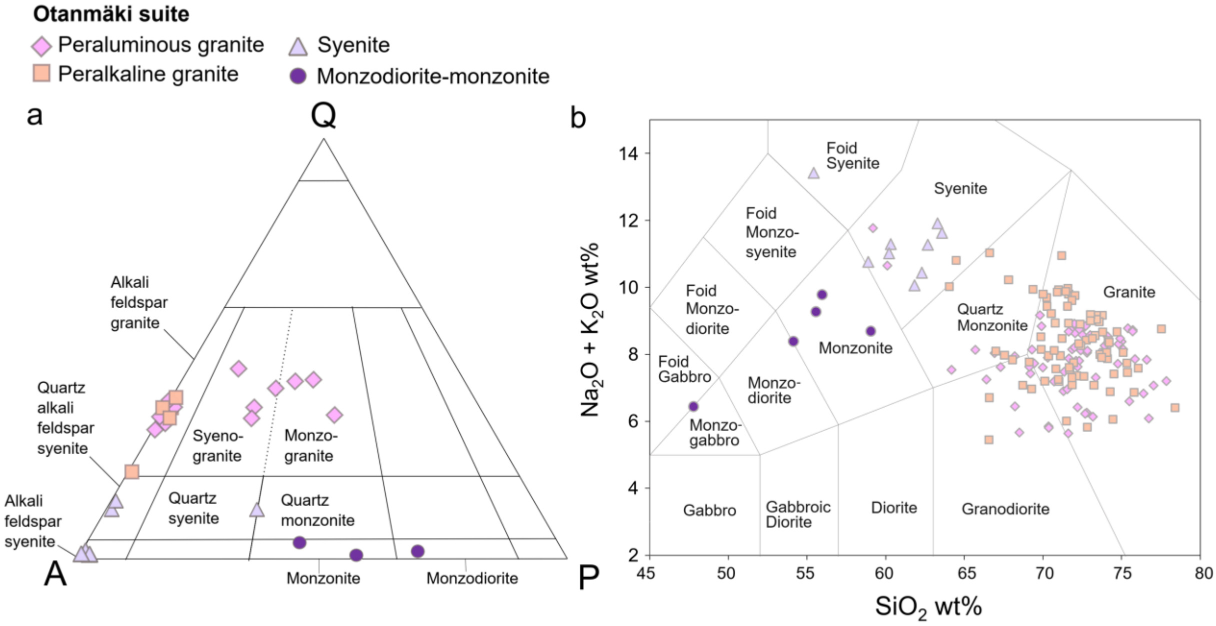

Figure 2. a) IUGS quartz-alkali feldspar-plagioclase (QAP) modal classification diagram (Streckeisen, 1967) and b) Total alkali vs. SiO$_2$ diagram (Middlemost, 1994) for Otanmäki suite felsic and intermediate igneous rocks (data from Papers I and II). Mineral abbreviations: Q = quartz, A = alkali feldspar (potassium feldspar, albite (An<5)), and P = plagioclase (An>5)

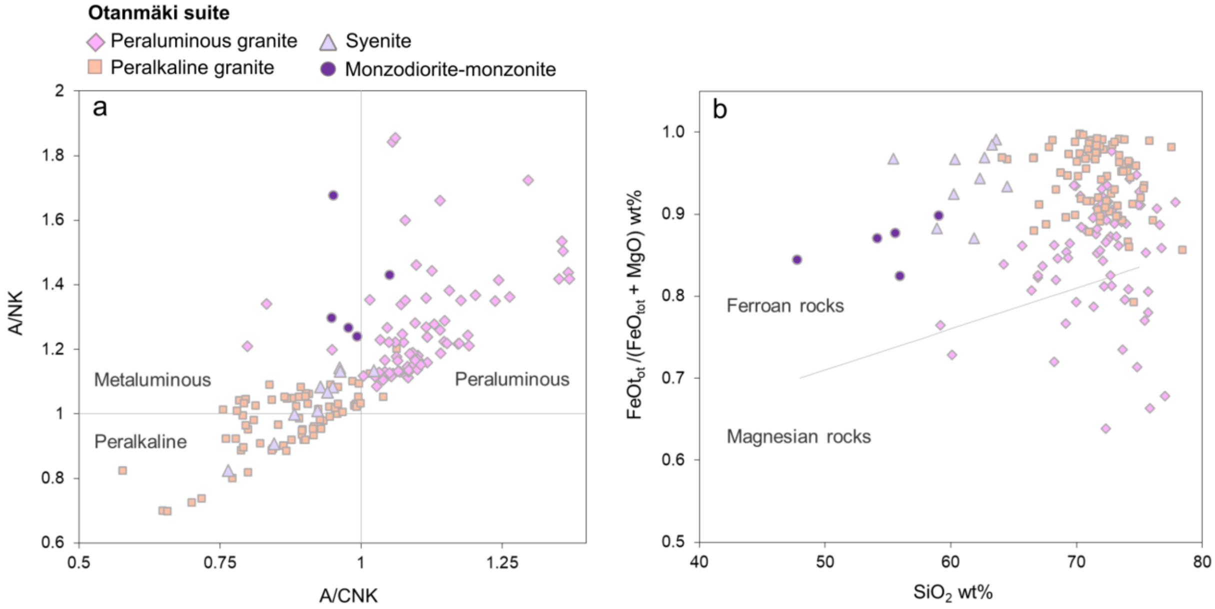

Figure 3. Major element classification diagrams for Otanmäki suite A1-type felsic and intermediate rocks. a) A/NK (molar Al$_2$O$_3$/(Na$_2$O+K$_2$O)) vs. A/CNK (molar Al$_2$O$_3$/(CaO+Na$_2$O+K$_2$O)) diagram (Shand, 1943). b) FeO$_{tot}$/(FeO$_{tot}$+MgO) vs. SiO$_2$ diagram (Frost and Frost, 2008. Data from Papers I and II

The eastern parts of the Otanmäki-Kuluntalahti nappe and the Lehtovaara lens are dominated by peraluminous granite (Fig. 1b), which occurs as coarse-grained plutonic and fine-grained, K-feldspar- and/or quartz-phyric subvolcanic types. Three samples of the coarse-grained type (A1896, A1832 and A1833) dated using CA-TIMS analysis yield probable magmatic U-Pb zircon ages of 2050 ± 2 Ma, 2046 ± 3 Ma and 2047 ± 3 Ma (respectively, Fig. 1b; Table 1). Zircon fractions from a sample of the fine-grained type (A1834, Fig. 1b; Table 1) were dated at 2047 ± 5 Ma, interpreted to represent the age of magmatic crystallization (CA-TIMS analysis; Table 1). The fine-grained type is observed to be frequently associated with quartzite-amphibolite-calc-silicate rock enclaves. Locally it intrudes with meandering contacts into a sequence of mainly "Jatuli-type" orthoquartzites with intercalations of mafic metavolcanic rocks and sills and also encloses several small inliers (<0.5 km^2), which contain calc-silicate rocks, intermediate gneisses, felsic volcanic breccias and conglomerates. A puzzling feature of the conglomerates is that they contain clasts of several rock types that are not known from the Jatuli and Kaleva sequences in the Kainuu belt or the Archean basement. These include pebble- to boulder-sized phenoclasts of alkaline intermediate to felsic plutonic and volcanic rocks. The narrowest central part of the main nappe is also fringed by a separate, 1x2-km-sized tectonic lens (Matojoki; Fig. 1b). It is composed of clast-supported rock-fall-avalanche conglomerates, which contain mostly gabbroic boulders but locally abundant felsic subvolcanic-volcanic rock boulders and cobbles as well.

Mafic intrusive rocks are scarce in the area, although the Otanmäki-Kuluntalahti nappe is in the western part fringed by foliated gabbroic bodies, which host Fe-Ti-V oxide ore bodies (Fig. 1b). These gabbro bodies are located along faulted contacts between the TTG gneisses and the Otanmäki suite rocks, having intrusive contacts with the former but not with the latter. The largest of them, the Otanmäki gabbro, has been dated at ca. 2.06 Ga (Huhma et al., 2018) and is thus roughly coeval with the ca. 2.05 Ga A1-type magmatism. In the west, the northern contact of the main nappe is lined by lenticular, highly strained gabbroic bodies of the Kemiläistenräme-Vuorijärvi mafic unit (Fig. 1b), whose correlation with the Otanmäki gabbro is possible but not yet ascertained.

The magmatic U-Pb zircon ages of 2.04–2.06 Ga obtained for the Otanmäki suite granitic members demonstrate that the rocks are pre-orogenic, having been formed ca. 150 Ma before the start of the Svecofennian orogeny, during which the plutons were metamorphozed and tectonized. The new U-Pb zircon data also demonstrate that the younger ages of ca. 1.96 Ga obtained from the Otanmäki suite granites record metamorphic re-equilibration effects on the ca. 2.05 Ga magmatic zircon grains.

The present map pattern and structural observations made across the Otanmäki suite nappes indicate that in the regional structural pattern, the nappes underwent major reshaping and rotation in the latest major deformation events of the Svecofennian orogeny. Because of the Svecofennian reworking and general poor exposure of contact zones, it is still unclear whether the granites and intermediate rock units in the western part of the main nappe had originally intrusive contacts or whether they represent intrusive bodies emplaced originally at different crustal levels but that were subsequently stacked together by displacements along faults. However, at least the peraluminous granite was emplaced at a shallow crustal depth as shown by the above-mentioned intrusive contacts with supracrustal rocks resembling Karelian cratonic cover strata. The A1-type rocks lack detected intrusive contacts with the surrounding Archean complexes and obvious Archean xenoliths, suggesting that the boundaries of the nappe units are indeed thrust faults involving a significant amount of displacement.

Microstructures of the A-type rocks are characterized by gneissic foliated and lineated fabrics in outcrops and microstructures indicative of a high degree of strain and mylonitization. The high-T deformation and metamorphism (above 550 ºC) make it difficult to judge how far (if not at all) the current minerals of the Otanmäki suite rocks represent their primary (igneous) mineralogy. However, in the modal QAP and total alkali–silica diagrams, the intermediate rocks and granites yield a very close correspondence in rock types (Figs. 2a and b). The monzodioritic-monzonitic rocks consist of calcic plagioclase, K-feldspar and variable proportions of amphibole, biotite, Fe-Ti oxides, and titanite. The syenitic rocks have QAP compositions of quartz syenite to alkali feldspar syenite and contain variable amounts of amphibole, clinopyroxene, biotite, and magnetite as mafic minerals. The peralkaline granite has a QAP composition of alkali feldspar granite and contains amphibole and/or clinopyroxene, and variable amounts of magnetite as mafic minerals. The QAP compositions of the peraluminous granite range from alkali feldspar granite to monzogranite, with minor amounts of biotite, amphibole and magnetite.

Geochemically, the granites and intermediate igneous rocks show a clear A-type character with a high Fe/Mg ratio (Fig. 3b) and high REE-HFSE and Ga contents (Fig. 4). More precisely, they belong to the A1-type of Eby (1992), which is interpreted to result from differentiation of OIB-like mantle-derived magmas. Global comparison to younger, mainly untectonized A-type suites shows that the Otanmäki suite has chemical characteristics similar to those of A1-type suites formed in intraplate hotspots and continental rifts by fractionation of variably crustally contaminated mantle-derived magmas. Igneous rocks similar in composition and age to the Otanmäki suite are not known from other parts of Finland, and even globally, A1-type rocks are rare in the early Paleoproterozoic geological record.

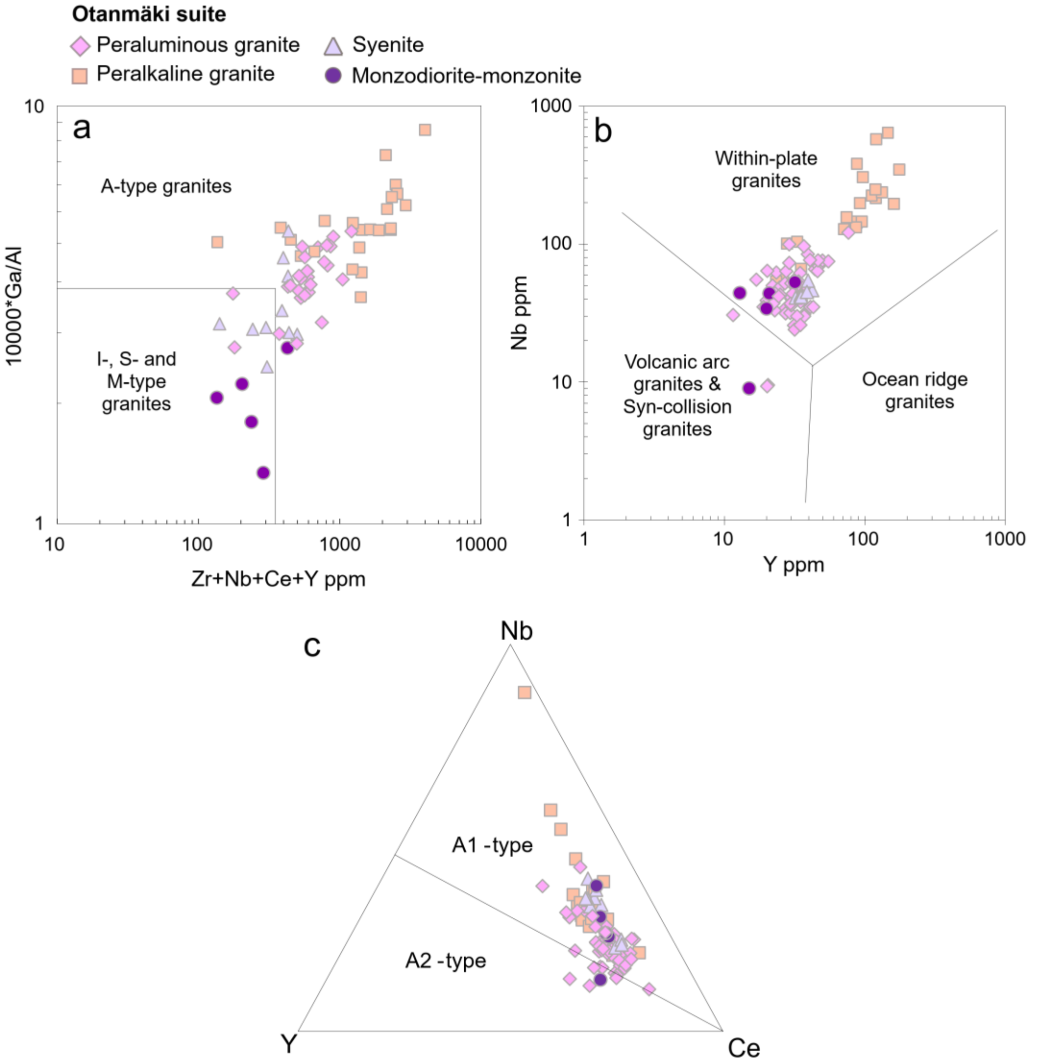

Figure 4. Tectonomagmatic discrimination diagrams for Otanmäki suite granites and intermediate rocks. a)-b) Trace element discrimination diagrams (Pearce et al., 1984; Whalen et al., 1987) for distinguishing A-type granites formed in within-plate settings from other types of granites. c) Ternary diagram of Eby (1992) discriminating between A1- and A2-type granite. Data from Paper I.

Based on its age, A1-type chemical affinity and present location, the ca. 2.05 Ga Otanmäki suite is interpreted to have emplaced during an early stage of a sluggish continental break-up event that occurred along the western margin of the Karelia craton and eventually led to the development of a continental passive margin and a nascent ocean basin, as witnessed by the 1.95 Ga Jormua ophiolite complex located within the same thrust belt as the Otanmäki suite nappes. The question of the original geologic setting of the Otanmäki suite is considered to remain debatable, but a parautochthonous nature of the Otanmäki suite nappes is favored based on the presence of metasedimentary rocks that superficially resemble the cratonic platformal and rift deposits found in the nearby Kainuu belt. An alternative model, in which the nappes would represent long-travelled rootless (exotic) bodies from the putative western margin of the Karelia craton, is not completely discarded because of the following reasons: 1) The apparently near complete lack of Otanmäki suite igneous rocks in the bedrock surrounding the nappes is easier to perceive in the exotic nappe model, 2) petrogenetic models predict that the A1-type granite genesis involved large volumes of temporally associated mafic rocks (cumulates) and the lack of physical or geophysical expressions of their presence in the nappes or surrounding bedrock is easily explained in the alternative model, and 3) the existence of metasupracrustal rocks that only seem to occur in the Otanmäki suite nappes also points to an exotic origin.

6.2 Paper II

Paper II presents results of a follow-up study of Paper I, focusing on detailed modeling of the magmatic evolution of the ca. 2.05 Ga Otanmäki suite A1-type peralkaline to peraluminous granites and intermediate igneous rocks (syenite, monzodiorite-monzonite). It reports new whole-rock geochemical and Sm-Nd isotope data for samples from the Otanmäki suite igneous rocks and results of thermodynamically constrained numerical modeling using the Magma Chamber Simulator (MCS) software equipped with the rhyolite-MELTS thermodynamic engine to derive phase equilibria. These data are applied to model fractional crystallization and assimilation processes (FC-AFC) inferred on geological and geochemical grounds. In addition, new whole-rock geochemical data are reported for gabbroic rocks and mafic dikes closely associated with the Otanmäki suite nappes.

Results of the new Sm-Nd isotope analyses demonstrate that the intermediate rocks show positive ε_{Nd}(2050 Ma) values (+1.3 to +2.6), which are only slightly lower than the modeled contemporaneous depleted mantle value (+3.4; DePaolo, 1981), but much higher than average ε_{Nd}(2050 Ma) of Archean TTGs (–10) in the surrounding bedrock (Fig. 5). This corroborates the view that the rocks of the Otanmäki suite were derived from a mantle-derived parental magma. The ε_{Nd}(2050 Ma) values of the peralkaline and peraluminous granite samples partly overlap with each other (–0.9 to +0.6 and –3.2 to +0.9, respectively; Fig. 5) and are somewhat lower than those of the intermediate rocks, suggesting that the mafic magmas parental to the granites likely have assimilated some amount of older Archean continental crust during their fractionation, which is consistent with the partly continental crust-like trace element signatures, such as Nb/U and Th/Nb ratios, of the granite members. It is established that mafic intrusive rocks that are closely associated with the Otanmäki suite nappes, but do not belong to the Otanmäki suite, can be divided into two types: 1) gabbroic intrusions and mafic dikes with a Fe-tholeiitic affinity and 2) alkaline mafic dikes with an OIB-like trace element affinity.

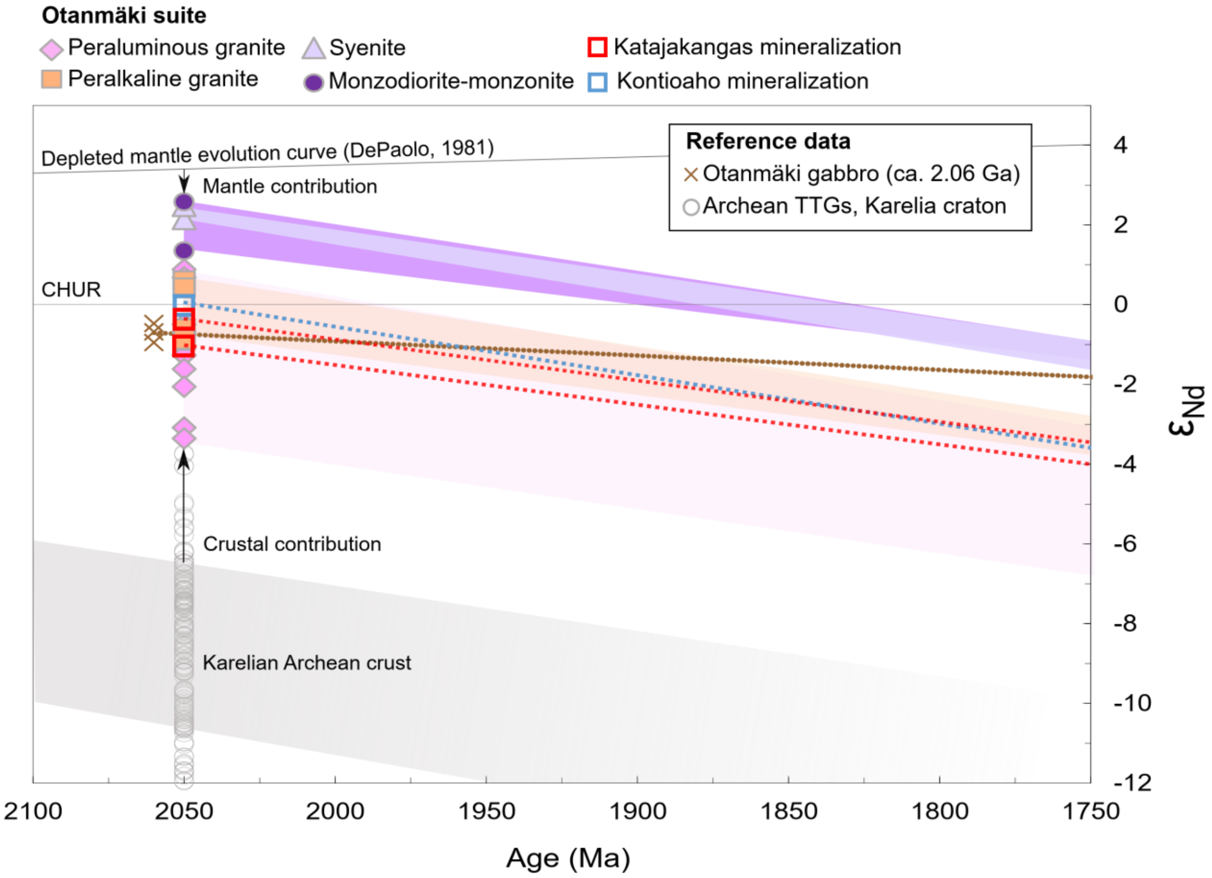

Figure 5. ε_{Nd} vs. age (Ma) diagram for samples of A1-type peralkaline and peraluminous granite and intermediate igneous rocks and Kontioaho and Katajakangas REE-HFSE mineralization from the Otanmäki suite (Data from Papers II and III). For comparison, data are also shown for samples from the ca. 2.06 Ga Otanmäki gabbro (Huhma et al., 2018) and Archean TTGs from the Karelia craton (ε_{Nd} calculated at 2050 Ma, Isotope database of Finland, www.gtk.hakku.fi/en). Depleted mantle evolution curve after DePaolo (1981).

Using MCS thermodynamic modeling of FC-AFC processes, it can be shown that a genetic connection is possible between the Otanmäki suite igneous rocks and the OIB-like alkaline mafic dikes from the Otanmäki area. Based on whole-rock compositions, two distinct types (I and II) of parental melt compositions are recognized. Their main difference is that type I has a higher Mg-number and MgO and CaO contents and lower TiO_2, Al_2O_3, Na_2O, P_2O_5, Nb, and Zr contents than type II, indicating that the former represents a more ‘primitive’ composition. The modeled FC paths are generally similar for both the type I and type II parental melts, especially in the early stage, involving extensive removal of clinopyroxene, olivine and plagioclase, producing a strong depletion in MgO and CaO and an increase in FeO_{tot}, SiO_2, Al_2O_3, Na_2O, K_2O and Fe-index in the resident magma. This induces stabilization of calcic plagioclase and minor amounts of olivine, Ti-rich spinel (i.e., titanomagnetite), apatite, and alkali feldspar, whereas the crystallization of clinopyroxene is suppressed. The late plagioclase fractionation results in a depletion in Al_2O_3 and CaO and an increase in SiO_2, Na_2O and K_2O in the resident magma, which becomes increasingly more peralkaline as the A/NK and A/CNK ratios in the magma decrease. This is known as the “plagioclase effect” (Bowen, 1945). The FC-only runs also demonstrate that the variation in the parental melt composition has a significant effect on the major element evolution and phase equilibria. The models run with the type I parental melt yield higher initial MgO and CaO contents, increasing the stability of clinopyroxene and olivine relative to that of plagioclase and spinel, whereas in the type II parental melt runs, the situation is the opposite, and the plagioclase effect is stronger.

It is established that FC involving extensive removal of olivine, clinopyroxene and calcic plagioclase from a mantle-derived OIB-like parental melt can explain the major element characteristics of the intermediate igneous rocks. The modeled melt compositions having the best-fit with the major element compositions of the Otanmäki suite intermediate rocks were produced by a high degree of fractionation (74 to 92%) of the type I parental melt in the 3–4 kbar pressure range, whereas the models using type II parental melt result in too low Al_2O_3. It is also noted that the compositions of the modeled late cumulates are compositionally similar to the intermediate rocks implying that they may well represent mixtures of cumulate minerals and residual magmas after FC.

Major and trace element, Nd isotopic and MCS modeling constraints suggest that the generation of the granites requires further fractional crystallization of OIB-like magmas coupled with assimilation of Archean TTGs. A tonalite sample from the study area, having a composition similar to that of the dominant low-HREE TTG component in the Karelia craton (e.g., Halla et al., 2009), provided the best fits in the AFC models and was therefore generally used. Compared to the FC-only models at the same pressure, the AFC models generate magmas with higher SiO_2, lower or similar Al_2O_3 and CaO and lower Na_2O and K_2O. The most important effect of assimilation is that it drives the residual melt composition to increasingly higher SiO_2 and increases the A/CNK and A/NK ratios, leading eventually to the crystallization of predominantly felsic minerals (alkali feldspars, quartz) instead of calcic plagioclase and mafic minerals. The AFC models demonstrate that melt compositions most similar to the Otanmäki suite granites require a high degree of fractionation of the parental melts (93–95%) in the 2.0–

2.5 kbar pressure range. The magma compositions generated using the type II parental melt correspond most closely to the Otanmäki suite peralkaline granite. This is mainly because the type II parental melt composition enables a high amount of plagioclase fractionation that functions against the increase in A/NK and A/CNK in the magma caused by assimilation of wall rocks, probably in form of wall-rock derived partial melts. In comparison, peraluminous silicic model melts similar to those of the Otanmäki suite are favorably produced from the type I parental melt AFC models, which only produces a moderate/weak plagioclase effect, resulting in relatively low-alkalinity residual magmas with a high aluminosity index. The AFC models suggest that just by varying the parental melt composition, while keeping the amount and composition of the assimilated crustal material relatively similar, the model melt trends approach both the peralkaline and peraluminous granitic compositions.

The outcomes of the MCS modelling show that magmas of A-type ferroan granites become more peraluminous by assimilation of quartzo-feldspathic crust, such as Archean TTGs. The AFC modeling results are also consistent with the idea that in addition to differing amounts of assimilated crustal melts, variations in the alkalinity and aluminosity in the end-product granite magmas can also depend on the differences in the parental melt composition, which affect the quantity of plagioclase fractionation, i.e., the plagioclase effect.

The FC-AFC models generated in Paper II predict the presence of complementary mafic cumulates with a volume of at least two times that of the related intermediate and granitic rocks. However, such rocks are not known to exist in the area or at surface or depth, strengthening the idea that the Otanmäki suite is part of a rootless allochthon.

6.3 Paper III

Paper III deals with the Otanmäki REE-HFSE mineralization hosted within the ca. 2.04–2.06 Ga Otanmäki suite granites. The main aim of the paper is to elucidate the timing and nature of the mineralizing processes and the source of the REE-HFSE using geological, geochemical and mineralogical data as well as Sm-Nd whole-rock and U-Pb zircon isotope data. The article also presents a synthesis of the extensive previous exploration data.

The Otanmäki REE-HFSE mineralization is contained in two occurrences, Kontioaho and Katajakangas, which are located within a fault-bound block composed mostly of ca. 2.06 Ga peraluminous A-type granite gneiss (Figs. 1b and 6a). The enclosing granite also contains slivers of metaturbidite and amphibolite metamorphosed from mafic dikes and showing shallowly (~10–30°) southwards dipping foliation and lineation (Fig. 6a). The peraluminous granite block has a shallow footwall contact against the adjacent peralkaline granite whereas its flanks appear to be controlled by near vertical faults against the surrounding peralkaline granite and syenite. The nearby peralkaline granite bodies contain local metaluminous parts.

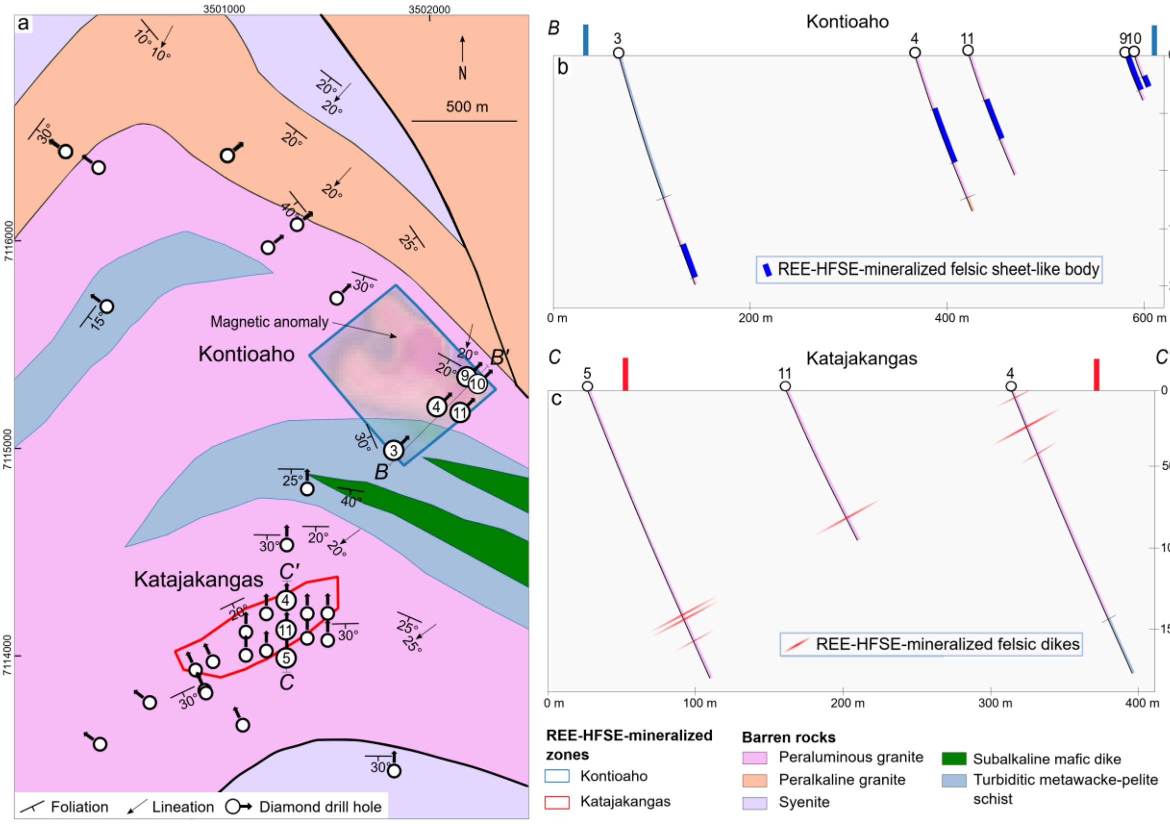

Figure 6. Local geology and occurrences of mineralized zones at Otanmäki. a) Geological map showing the inferred surface projections of the Kontioaho and Katajakangas mineralized zones and diamond drill holes. b–c) Simplified vertical cross-sections of the mineralized rock units (modified after Paper III). The drilling profiles (B–B’, C–C’) shown in Fig. 6a. Coordinates in KKJ-3/Finland Uniform Coordinate System. Published with permission of Bulletin of the Geological Society of Finland.

Table 2 Minerals found in the Kontioaho and Katajakangas REE-HFSE-mineralized zones together with their structural formulae and abundances (wt% values) in representative samples (Paper III).

Sample		KKNOK (outcrop)	KK-R5-152.70m	KA-R3-196.26m	KA-MD-6 (outcrop)
Mineral	Structural formula	Katajakangas mineralized dikes		Kontioaho mineralization	
Gangue minerals					
Quartz	SiO_2	59	61.4	48	36
Potassium feldspar	$KAlSi_3O_8$	0.4	0.3	25.4	19.5
Albite	$NaAlSi_3O_8$	5.9	6.2	2.3	26.8
Magnetite	Fe_2O_4	*nd*	*nd*	4.6	6.6
Fluorite	CaF_2	*nd*	*nd*	1.9	0.5
Calcite	$CaCO_3$	1.5	3.4	2.1	0.1
Biotite	$K(Mg,Fe)_3(AlSi_3O_{10})(F,OH)_2$	0.7	0.9	0.2	0.4
Chlorite	$(Mg,Fe)_3(Si,Al)_4O_{10}(OH)_2(Mg,Fe)_3(OH)_6$	0.1	0.1	0.3	0.1
Amphibole	$(Na,Ca)_2(Fe^{2+}{}_4Fe^{3+})(Al_2Si_6O_{22})(OH)_2$	0.2	0.2	0.7	0.4
Andradite	$Ca_3Fe_2Si_3O_{12}$	0.2	0.2	0.1	1.2
Hydroxylapatite	$Ca_5(PO_4)_3(F,OH)$	0.5	0.6	0.1	0.2
Pyrite	FeS_2	2.5	2.5	*nd*	*nd*
REE-HFSE-bearing minerals					
Allanite-(Ce)	$(^{*}Ln,Ca,Y)_2(Al,Fe^{2+,3+})_3(SiO_4)_3(OH)$	19.5	14.7	4.4	2.8
Zircon	$(Zr,Hf,Ln,Y,Nb,U,Th)SiO_4$	2.3	2.5	5.5	2
Titanite	$(Ca,Y,Ln)(Nb,Ti,Si)O_5$	0.8	1.2	1.5	1.5
Nb-REE-Th-U oxides		2.8	4.3	0.6	0.3
Fergusonite-(Y)	$(Y,Ln,U,Th)NbO_4$				
Samarskite-(Y)	$(Y,Ln,Fe,U,Th,Ca)(Nb,Ta)_5O_4$				
Euxenite-(Y)	$(Y,Ca,Ln,U,Th)(Nb,Ta,Ti)_2O_6$				
Aeschynite group	$(Y,Ln,Ca,Fe,U,Th)Ti,Nb)_2O_6$				
Pyrochlore group	$(Ca,Na,Y,Ln,U,Th)_2Nb_2O_6(OH,F)$				
Columbite-(Fe)	$Fe^{2+}Nb_2O_6$				
Fersmite	$(Ca,Ln,Na)(Nb,Ta,Ti)_2(O,OH,F)_6$				
REE fluorocarbonates		0.4	0.5	0.6	0.4
Parisite-(Ce)	$Ca(Ln)_2(CO_3)_3F_2$				
Bastnäsite-(Ce)	$(Ln)CO_3F$				

*Ln = lanthanides; *nd* = not detected. Weight proportions of minerals determined by FE-SEM based automated mineralogy system (Mineral Liberation Analyzer; MLA) from polished thin sections.

Both mineralized zones have a minimal outcrop coverage, are outlined only by scarce diamond drilling, and their strike and depth extensions remain open. The mineralized rocks are fine-grained and show gneissic structures and sharp contacts against their wall rocks. At Kontioaho, the mineralization occurs as a 30– to 50-m-thick felsic sheet-like body, with a drilling-tested depth extent of 185 m and a strike length of only a 100 m (Figs. 6a and b). The Kontioaho mineralization is magnetite-bearing and consequently causes a magnetic anomaly, which suggests that it extends approximately 600 m to NW from the existing drilling profile (Fig. 6a). At Katajakangas, the mineralization consists of several 0.1– to 1.4-m-thick felsic dikes, restricted to a ca. 100-m-wide, tabular zone, which is traced to a depth of 145 m with a strike extension of at least 800 m (Figs. 6a and c).

Gangue minerals in the mineralized rock units include quartz, potassium feldspar, albite, and in varying proportions magnetite, fluorite, calcite and pyrite (Table 2). Miaskitic mineral assemblages comprising allanite-(Ce), zircon, titanite, and Nb-REE-Th-U oxides characterize both mineralization (Table 2). Allanite-(Ce) is recognized as the major host for REEs and Th in both occurrences, being rich in LREE (La-Sm). Zircon is the only major host for Zr and a minor host for Th, U, Nb, Y, and heavy REEs (Gd-Yb). A variety of Nb-REE-Th-U-rich oxide minerals (e.g., fergusonite-(Y), samarskite-(Y), pyrochlore) are the major carriers of Nb, Y, heavy REEs, Th, and U in the Katajakangas dikes, but are less abundant in the Kontioaho sheet-like body. Titanite is an important carrier of Nb, in addition to being a minor carrier of REEs in the Kontioaho body.

In-situ SIMS U-Pb dating conducted on zircon grains from the Kontioaho REE-HFSE mineralization yield an average $^{207}Pb/^{206}Pb$ age of 2036 ± 4 Ma, which is (within error) close to the crystallization age of the peralkaline granite in the Otanmäki area (ca. 2.04–2.05 Ga; Fig. 1b; Table 1). Altered domains in the analyzed zircon grains from Kontioaho yield younger U-Pb ages of ca. 1.9 Ga, which coincide with the initiation of the Svecofennian orogeny and regional metamorphism, demonstrating effects of metamorphic overprint.

The mineralized rocks both at Kontioaho and Katajakangas exhibit SiO_2 contents that are typical of granitic rocks (~66–71 wt% and ~68–82 wt%, respectively), although some samples with high calcite and/or fluorite contents show lower SiO_2. A geochemical affinity of the mineralized rocks to the peralkaline granite in the Otanmäki area is shown in primitive mantle-normalized spidergrams, in which all mineralized rocks display pronounced depletions in Ba, Sr, Eu, P, Ti relative to REEs (except Eu), Nb-Ta, Th-U, and Zr-Hf. Also, compared to the peralkaline granite, the mineralized rocks at Kontioaho and Katajakangas show similarly high

Fe/Mg ($FeO_{tot}/(FeO_{tot}$ + MgO) = ~0.8–1.0) and enrichment in LREE relative to HREE (LREE/HREE = ~3–6) and similarly negative Eu anomalies with Eu/Eu* of ~0.3–0.5. However, they differ from the peralkaline granite in having higher REE-HFSE and CaO and lower Al_2O_3 and total alkalies, and based on their alkalinity, the mineralized samples can be classified as metaluminous. In terms of REE-HFSE contents, the Katajakangas dikes show the most extreme compositions, except for Zr, which displays the highest concentration in the mineralized rocks at Kontioaho. Fluorine correlates well with REE-HFSE and CaO in the Kontioaho mineralized body, but in contrast, the Katajakangas mineralized dikes have low contents of F. Instead, CO_2 and S are high in some mineralized dikes at Katajakangas.

Neodymium isotope compositions measured for the peralkaline granite and peraluminous granite wall rock give ε_{Nd}(2050 Ma) values of –0.9 to +0.6 to and –1.3, respectively and the ε_{Nd}(2050 Ma) values (–1.1 to 0.0) of mineralized samples are comparable to those of the peralkaline granite (Fig. 5). These values suggest derivation of all these rocks essentially from a mantle source with relatively minor contributions to their magmas from an older, highly fractionated material, such as TTGs in the Archean basement.

The formation of the REE-HFSE enrichment is explained by a multistep process, which involved generation of highly evolved silicate melts during the Otanmäki suite A1-type magmatism. The peraluminous monzogranite wall rock is interpreted to be genetically unrelated to the mineralized rock units, as evidenced by whole-rock chemical and Sm-Nd isotope data and zircon U-Pb geochronology. Instead, the mineralization is considered to be intrinsically related to the crystallization history of the ca. 2.04–2.05 Ga peralkaline granite magmatism of the area, particularly later stages in its evolution. The peralkaline granite related to mineralization is proposed to have crystallized from an evolved peralkaline felsic magma with high initial REE-HFSE content. The generation of the wt%-level enrichment of REE-HFSE in the Otanmäki area required extensive crystallization of this magma in order to accumulate REE-HFSE to the last residues of the silicate melt.

Based on the performed numerical modeling, it is established that extensive crystallization of alkali feldspar, quartz, and minor mafic minerals (aegirine, magnetite) from a peralkaline granitic starting magma composition can produce a residue with major element compositions (e.g., Si, Fe, and Ca enrichment relative to Al, K, and Na) similar to those of the Otanmäki REE-HFSE mineralized rock units, although the observed Ca enrichment likely required an additional process. Trace element modeling also shows that pure fractionation alone cannot be responsible of the extremely high REE-HFSE levels in the mineralization. Based on elevated

F, CO_2 and S contents in the mineralized rocks units, it is suggested that further REE-HFSE and Ca enrichment took place due to complexing of REE, HFSE and Ca with volatile components (e.g., F^-, CO_3^{2-}, SO_4^{2-}), which strongly sequestered these metals to the very late-stage melts. The fluorine-dominant composition of the volatile phase at Kontioaho appears to have favored enrichment of Zr relative to Nb and REEs. At Katajakangas, the mineralized felsic dikes are more enriched in Nb and REE relative to Zr, and the volatile phase appears to have been more enriched in CO_2 and S relative to F. In addition, the Eu/Eu* ratio, which exhibits only a slight decrease in mineralized rocks compared to the peralkaline granites, points to an oxidized nature of the late-stage melts.

The above-mentioned processes are considered to have produced metaluminous, high-silica residual melts strongly enriched in REE-HFSE, Ca, and Fe relative to Na, K and Al from a peralkaline felsic parental melt. The final step in the process was accumulation of these last residues of silicate melt and their emplacement as highly fractionated dikes and a sheet-like intrusive body. The allanite-zircon-titanite-Nb-REE-Th-U oxide dominant REE-HFSE mineral assemblages evidently record some textural and compositional changes related to metamorphic re-equilibration, but the main mineral species/assemblages are interpreted to be essentially primary, reflecting the magmatic crystallization conditions in a relatively low-alkalinity (metaluminous), oxidized, Ca-, Si-, Fe-, Al-rich and P-poor and low-fCO_2 system.

7 Discussion

7.1 Tectonic setting of the Otanmäki suite

The Otanmäki suite A1-type magmatism records a previously poorly documented episode of ca. 2.05 Ga continental rifting-related intermediate to felsic magmatism with strong links to contemporaneous OIB-like mantle-derived magmatism (Papers I and II). However, the suite remains enigmatic as to why there is a complete absence of similar rocks elsewhere in the preserved rock record of the western margin of the Karelia craton (Paper I).

In order to understand the spatial and temporal position of the Otanmäki suite magmatism during the rifting and break-up of the Karelia craton, the post-Archean evolution of the craton, must be first examined. The time period from ca. 2.45 to 2.10 Ga was an era of relative tectonic quiescence with minimal or no record of accretional/collisional processes (Mikkola et al., 2011; Hanski and Melezhik, 2013b; Nironen, 2017). During this period, there was significant erosion and peneplanization of the Karelia continent but also several discrete episodes of crustal extension resulting in opening of intracontinental rift basins (Laajoki, 2005; Vuollo and Huhma, 2005; Hanski and Melezhik, 2013a,b: Lahtinen et al., 2015). The Paleoproterozoic rifting episodes of the Karelia craton were associated with mafic magmatism involving repeated events of mafic dike swarm formation (and associated shallow level sills) and emplacement of intrusions and extrusive rocks at ca. 2.45, 2.4, 2.2, 2.15, 2.12, and 2.10 Ga, but it is unlikely that all these extensional/rifting episodes were related to true continental break-ups (e.g., Lahtinen et al., 2010, 2015; Nironen, 2017; Davey et al., 2020). The ca. 2.45–2.4 Ga mafic magmatism was also associated with small volumes of granite magmatism in northern Finland (Lauri et al., 2012).

The 2.1–2.05 Ga stage involved major rifting of the Karelia continent, which eventually led to its break-up along its present western margin (Lahtinen et al., 2010, 2015; Nironen, 2017). At ca. 2.1 Ga, large areas of the Karelia craton were affected by voluminous mafic magmatism as exemplified by the widespread occurrence of ca. 2.1 Ga Fe-tholeiitic dike swarms and shallow level sills in eastern Finland and adjacent Russia. All this magmatism is thought to be connected to a 2.1 Ga mantle plume event (Vuollo and Huhma, 2005; Hanski, 2013; Huhma et al., 2018; Davey et al., 2020). The craton-wide 2.1 Ga mafic magmatism was also associated with small volumes of granite magmatism in northern Finland (Ahtonen et al., 2007). Mafic volcanic rocks and intrusions having an age of ca. 2.05–2.06 Ga exist in northern and central Finland (Perttunen and Vaasjoki, 2001; Perttunen and Hanski, 2003; Hanski and Huhma, 2005; Lukkarinen, 2008; Huhma et al., 2018), although the volume of mafic magmatism at this time was probably less than at 2.1 Ga. Felsic magmatism at ca. 2.05 Ga is scarce as in addition to the Otanmäki suite, there are only two small occurrences of felsic subvolcanic and effusive rocks of this age in the Paleoproterozoic supracrustal rocks in the Peräpohja belt (Perttunen and Vaasjoki, 2001) and Siilinjärvi area (Lukkarinen, 2008). Elsewhere in the Fennoscandian Shield, similarly aged alkaline mafic to felsic volcanic rocks exists in the Pechenga belt in the Kola Peninsula, NW Russia (Melezhik and Hanski, 2013). The Pechenga belt is interpreted to have formed by rifting and continental break-up that took place between the Karelia craton and the Kola craton (Daly et al., 2006).

The Karelian formations in eastern Finland comprise mainly metasedimentary rocks, which are subdivided into 1) autochthonous 2.5 to 2.0 Ga aeolian to transgressive fluvial and deltaic shallow-water deposits on the Archean basement (Sariola–Jatuli) and 2) younger 1.92 to 2.0 Ga Kaleva strata consisting of autochtonous–parautochthonous and allochthonous, deeper water deposits, which were thrust from the west onto the Archean basement complex (Laajoki, 2005;

Lahtinen et al., 2010; 2015). The provenance of the detrital material in the Lower Kaleva metasediments was in the deeply weathered Archean basement and its pre-Kaleva sedimentary cover and are interpreted to reflect sedimentation after ca. 2.06 Ga in a continental margin rift basin devoid of syn-rift magmatism (Lahtinen et al., 2010; Kontinen and Hanski, 2015). The Upper Kaleva system metasediments are excluded from the continental rift fill as they contain a large Palaeoproterozoic detrital component with maximum depositional ages between 1.92 to 1.95 Ga (Lahtinen et al., 2010).

The Karelia craton in Finland and Russia also records NW-trending 1.95–1.98 Ga mafic dikes (Vuollo and Huhma, 2005; Lubnina et al., 2016; Huhma et al., 2018) and ca. 1.98 Ga flood basalts in Russian Karelia (Puchtel et al., 1998). At 1.95–1.96 Ga, the crustal units of the future Jormua and Outokumpu ophiolites were formed. The Jormua complex comprises a well-preserved ophiolitic sequence consisting of Archean subcontinental lithospheric mantle rocks accompanied by ca. 1.95 Ga gabbros, sheeted dike complexes and pillow lavas (Kontinen, 1987; Tsuru et al., 2000; Peltonen, 2005; Peltonen et al., 2008). The Outokumpu ophiolite is more restricted in its rock types, consisting of numerous mantle tectonite massifs without associated sheeted dikes or abundant extrusive rocks (Peltonen et al., 2008). Proto-ophiolites for the Jormua and Outokumpu ophiolitic fragments are considered to have formed in a setting involving final break-up/ocean opening in an essentially magma-poor tectonic environment characterized by asymmetric extension (Peltonen and Kontinen, 2004; Peltonen, 2005). The Jormua ophiolite is thought to have developed in the initially opened part of a rift basin and the Outokumpu ophiolite closer to the continental margin at ca. 1.95 Ga (Peltonen, 2005; Peltonen et al., 2008; Huhma et al., 2018).

The above-discussed evidence from the rock record of the western margin of the Karelia craton is consistent with events of rifting and lithosphere thinning from 2.1 to 1.95 Ga, but it is still debatable how the actual craton breakup took place. The scarcity of ca. 1.95 Ga volcanic activity in the rock record at the craton margin made Peltonen and Kontinen (2004) to propose a model in which the final break-up/ocean opening took place at ca. 1.95 Ga in a magma-poor margin. Because modern magma-poor margins are associated with very slow spreading rates, Lahtinen et al., (2010, 2015) argued that any wide ocean could not have formed and closed in the time span of less than 50 Ma available in the 1.95 Ga break-up scenario. Instead, they suggested a model in which the western Karelia margin accommodated both magma-rich and magma-poor margins. Lahtinen et al. (2010) suggested that the onset of break-up was marked by bimodal magmatism at ca. 2.05–2.06 Ga and voluminous deposition of turbidites in rift basins and along the continental margin. The magmatism in the passive margin is suggested to have localized in "soft points" formed at locations where rifts were offset by transform faults. Continental break-up is suggested to have occurred at 2.05–2.1 Ga followed by new spreading within the pre-existing margin at ca. 1.93–1.98 Ga in a non-volcanic margin setting (Lahtinen et al., 2015).

A problem with the models of evolution of the Karelia break-up is that most of the evidence that has been used to characterize the break-ups and the thinned margin comes from allochthonous units thrust over the Archean craton, which may have their tectonic provenance/roots in the once existed but now drifted-away part of the craton. In such a case, the sample record of the actual margin could be very scanty and biased. The present Svecofennia-Karelia boundary zone is dominated by crustal-scale strike-slip tectonics (Lahtinen et al., 2015), which may have removed the thinned continental crust and exposed lithospheric mantle already before the collision of the Svecofennian domain. This interpretation is also supported by the results of Papers I and II, which favor the view that the Otanmäki suite nappes represent rare thrust units, analogous to the Jormua-Outokumpu allochthon consisting of ophiolite fragments and Upper Kaleva metasediments, whose provenance was in a nascent ocean west of the putative lost continental margin. Compared to the Jormua-Outokumpu allochthon, the Otanmäki suite nappes appear to represent rare thrust samples from more inland parts of the since 2.05 Ga created rifted margin (Papers I and II). The inland nature of the Otanmäki suite is shown by their geochemistry and proposed modes of origin, which required fractionation of mantle-derived OIB-like parental melts and their interaction with TTG-series gneisses at relatively shallow crustal depths (~10 km) (Paper II) (Fig. 7a).

The development of asymmetric rifting/extension along low-angle lithospheric detachment faults reaching the base of the continental crust as well as thermal subsidence of the Karelia craton have been previously suggested as an explanation to the formation of the depositional basins for the Paleoproterozoic cover rocks deposited between ca. 2.4 and 2.1 Ga (Strand, 1993; Lahtinen et al., 2010) and subsequent exhumation of the subcontinental lithospheric mantle upon development of a passive continental margin at ca. 1.95 Ga, as recorded by the Jormua and Outokumpu ophiolites (Kontinen, 1987; Peltonen, 2005). The detachment structures and related arrays of faults generated in association with continental rifting and break-up (cf. Stockli and Bosworth, 2018) provide pathways for mantle-derived melts, such as those suggested to be parental to the Otanmäki suite (Paper II). Some of the Otanmäki suite A1-type magmas appear to have reached the uppermost parts of the crust as the peraluminous granite in the Otanmäki suite nappes have intrusive contacts with Paleoproterozoic metasediments, which likely represent rocks detached from pre-rift cratonic and rift sequences in the putative, pre-1.9 Ga craton margin (Paper I). The transport distances of the Otanmäki suite nappes remain unknown, but the fault-defined nature of the contacts between the A1-type rocks and surrounding Archean rocks and the lack of detected intrusive contacts between them also support the interpretation that the nappes containing the A1-type rocks have experienced a considerable amount of displacement along the faulted contacts (Paper I). It also remains enigmatic what was the temporal and spatial relationship between the ca. 2.05 Ga A1-type rocks and the ca. 2.06 Ga Fe-Ti-V oxide-bearing gabbro intrusions in the the Otanmäki area as these rocks are separated by faults with no evidence of intervening, although the gabbros are locally intruded by meter-wide A-type granite dikes (Papers I-II). Dating of these granite dikes could provide more information on the temporal and spatial relationship between mafic and felsic magmatism in the Otanmäki area and should therefore be included in the future research plans (Paper I).

In conclusion, the general rarity of the ca. 2.05 Ga A1-type igneous rocks represented by the Otanmäki suite igneous rocks can be explained by the notion that they constitute long-travelled allochthonous sheets thrust from a distal, now disappeared part of the Karelia margin, the "lost or hidden west", which possibly hosted more similar ca. 2.05 Ga A1-type intrusions. The tectonic provenance and original geodynamic setting of the Otanmäki suite nappes remain open to discussion but fortunately, there are good opportunities to acquire more critical data, including depositional ages and sources of metasupracrustal rocks enclosed in the Otanmäki suite nappes. Preliminary investigations have already shown that these units contain conglomerate occurrences with phenoclasts, which record a 2.03–2.05 Ga felsic volcanic-sedimentary event previously unrecognized from the western margin of the Karelia craton (Kärenlampi et al., 2021). Clearly, further studies of the Otanmäki nappes are likely to produce more valuable information on the 2.05 Ga tectonic processes, which are needed for advancing our understanding of the break-up event of the Karelia craton.

7.2 Origin of the Otanmäki suite A1-type igneous rocks

The presented geochemical and Nd isotopic data and numerical modeling using Magma Chamber Simulator in Paper II provide a good foundation for deciphering the sources and petrogenetic pathways to igneous suites showing a wide variety of A-type ferroan granitic and intermediate igneous rocks emplaced in the same igneous complex.

The A1-type granite trace element signature and OIB-like affinity of the Otanmäki suite igneous rocks, their mantle-like Nd isotope compositions, and constraints obtained by thermodynamic

FC-AFC modeling using the MCS show that they can be plausibly produced from OIB-like mafic parental melts (Paper II). These results also demonstrate that the primary requirement for the generation of the Otanmäki suite magmatism was the availability of OIB-like parental basaltic magmas.

Ocean island basalts and OIB-like basalts are widespread both in oceanic and continental settings (e.g., Fitton, 2007). OIBs are commonly olivine-bearing lavas with subalkaline (tholeiitic) and alkaline compositions (e.g., Suetsugu et al., 2005). Alkali basalt is the most abundant type in alkaline OIB suites, but more alkaline lavas, such as basanite and nephelinite, also commonly erupt in many ocean islands as well as continental settings (Fitton, 2007). Compared with mid-ocean ridge basalts (MORB), both tholeiitic and alkalic OIBs have higher concentrations of incompatible trace elements, and their chondrite-normalized rare earth element patterns show strong enrichment in light REEs relative to heavy REEs (e.g., $(La/Sm)_{CN}>1$). Alkaline OIBs are typically more enriched in incompatible elements, including light REEs, compared to tholeiitic OIBs (Suetsugu et al., 2005; Guo et al., 2019).

In general, OIBs, both tholeiitic and alkaline, are thought to originate from deep mantle sources, with melting beginning in the stability field of garnet peridotite, imparting the familiar 'garnet signature' to all OIB melts (e.g., $(Sm/Yb)_{CN}>1$), an inference that places the onset of melting at depths greater than about 70 km (e.g., Niu et al., 1999; Putirka, 1999). Moreover, variation in the lithosphere thickness has been suggested to be the primary control on OIB geochemistry on a global scale, both in continental and oceanic settings (Niu et al., 2011; Guo et al., 2019). Alkaline OIB magmas are produced in areas of a thick lithosphere and have geochemical signatures consistent with a low-degree and high-pressure of partial melting or final melt equilibration. In contrast, basalts erupted on a thin lithosphere, the intensity of the garnet signature decreases with increasing extent of melting, which takes place at lower pressure in the spinel peridotite facies (Niu et al., 2011; Guo et al., 2019). The OIB-like mafic dikes used as a proxy for the parental melt composition for the Otanmäki suite A1-type rocks show a low-degree and deep-pressure melting signature (i.e., are highly enriched in incompatible elements, such as Zr, Nb, REE, and show the 'garnet' signature with high $(La/Sm)_{CN}$ (~5) and $(Sm/Yb)_{CN}$ (~4); Paper II), indicating their origin below a thick lithosphere.

The generation of such a variability in igneous rock types as observed in the Otanmäki suite requires that batches of the OIB-like parental magmas evolved along different differentiation paths. Based on trace element signatures and MCS modeling results (Paper II), the origin of the Otanmäki suite intermediate igneous rocks (monzodiorite-monzonite, syenite) required that their OIB-like parental melts evolved by extensive fractional crystallization at relatively shallow depths in the middle to upper crust (~2–4 kbar, ~7–15 km), in a process that involved crystallization of large amounts of clinopyroxene, plagioclase and olivine (Fig. 7a). Paper II also highlights the importance of crustal assimilation in the generation of the Otanmäki suite granites, in the form of fractional crystallization combined with assimilation of Archean wall rocks, with typical TTG-series gneisses providing a well-fit assimilant (Paper II) (Fig. 7a). Assimilation of partial melts from protoliths such as TTGs are shown to turn the end-product granite magmas increasingly more felsic and peraluminous, supporting the previous perceptions on ferroan A-type granite genesis (see Chapter 2.3). However, the results of Paper II also refine the previous propositions by showing that differences in the initial compositions of the parental melt undergoing FC and AFC also have a significant effect on the differentiation paths and final products of the differentiation process. Above all, differences in the parental melt major element composition affect the quantity of late plagioclase fractionation, which explains the variation in the alkalinity and aluminosity of ferroan A-type granites, even in cases where there were similar amounts of crustal assimilation. In addition, it is shown that the extensive differentiation required in the Otanmäki suite petrogenesis involved closed-system conditions where replenishment and tapping events were prevented as these processes would effectively inhibit the generation of highly fractionated magmas, instead resulting in nearer to steady-state (more mafic) compositions (Paper II) (Fig. 7a).

The results of Papers I-III demonstrate that compositionally sufficiently variable igneous complexes, even when metamorphosed under high-T conditions, can still yield workable information on the origin and evolutionary histories of ferroan A-type intermediate to felsic magmas, to the extent of providing a basis for understanding the link between continental OIB-like mafic magmatism and intermediate to felsic igneous rocks emplaced closely in time and space within the same igneous complex. However, several important petrogenetic questions are still to be answered in the Otanmäki area. For example, what were the original (pre-orogenic) spatial and temporal relationships and emplacement depths of the peralkaline and peraluminous granites and the intermediate igneous rocks, and why the intermediate rocks seem to lack significant crustal assimilation? There is a good potential to obtain further information on the source characteristics of the Otanmäki suite igneous rocks and their petrogenetic pathways. The pre-existing geological, geochemical and U-Pb and Sm-Nd isotope data generated in Papers I-III create a good basis for follow-up studies using, for example, other isotope methods (e.g., zircon Lu-Hf and O isotopes) to further clarify the source characteristics of the Otanmäki suite igneous rocks. From a petrogenetic point of view, it would also be interesting to compare the mineralogy and mineral chemistry of the Otanmäki suite to those of chemically similar but unmetamorphosed and undeformed igneous rocks (e.g., Vilalva and Vlach, 2014; Siegel et al., 2017b) to find out if the compositions of the minerals in the Otanmäki suite rocks could still yield information of the conditions of their magmatic crystallization. In a positive case, mineral chemistry could also be utilized to obtain more information on the REE-HFSE mineralization processes (see Chapter 7.3).

Although the results of Paper II demonstrate that the genesis of ferroan A-type granites of varying alkalinity and aluminosity, and that of related intermediate igneous rocks, can be explained largely by assimilation and fractional crystallization, it must be kept in mind that the study concerns the genesis of A1-type granites, which generally show strong genetic links to contemporaneous OIB-like mafic magmatism, and cannot be applied to all A-type granites. For example, the ferroan A2-type granite suites, which are globally much more common and more voluminous than the A1-type suites (e.g., Bonin, 2007; Frost and Frost, 2013; Grebennikov, 2014), show a strong continental fingerprint in their compositions, which points to a significant role of crustal melting in their genesis, rather than to an origin by FC-AFC of mantle-derived parental melts. On the whole, A-type granites come in many flavors with different modes of origin and therefore their petrogenesis is a wide field of research (see Chapter 2).

7.3 Origin of the REE-HFSE mineralization at Otanmäki

In the preceding chapters, various processes that contributed to the formation of A1-type granitic magmas during the rifting of the Karelia craton were examined and discussed. However, these processes alone do not explain the conditions and processes resulting in the high REE and HFSE concentrations of several wt% in the known mineralization at Otanmäki, but further enrichment of these elements was required (Paper III) (Fig. 7b).

The first step in the mineralization process was generation of a batch of peralkaline granite magma (Paper III), which, as based on Paper II, involved low-degree partial melting of an enriched OIB-like mantle source followed by extensive magmatic differentiation of the magma by removal of a plagioclase-dominant mineral assemblage at a relatively low pressure (~2 kbar). A similar origin has been proposed for many other REE-HFSE-rich peralkaline granites (e.g., Vasyukova and Williams-Jones, 2020). In the Otanmäki case, assimilation of continental crust is an important additional factor that explains how a basaltic parental melt evolved to granitic compositions (Paper II). Significant part of the assimilation likely occurred in non-replenishing magma chambers at upper crustal depths and involved assimilation of partial melts from enclosing TTG-series gneisses (Paper II). The total amount of assimilated crust in the most evolved peralkaline granites is estimated to be 30% of the initial mass of the mafic parental melt (Paper II). Assimilation appears to have been essential in the generation of the REE-HFSE-rich peralkaline felsic melts as it turned the residual magma increasingly more felsic in compositions and lower in its solidus temperature, allowing a prolonged process of fractional crystallization and generation of REE-HFSE-rich peralkaline silica-saturated products (Paper II). Crustal assimilation likely also increased the overall REE-HFSE budget, although the principal source of REE-HFSE was ultimately in the mantle. The MCS trace element and Nd isotope modeling (Paper II) indicated that 2% of Nb, 27% of Nd, and 50% of U in the best-modeled peralkaline granitic melt can be attributed to crustal assimilation.

The next step in the process of REE-HFSE enrichment at Otanmäki involved extreme fractional crystallization of peralkaline magma elevating the REE-HFSE contents to wt% levels in the last residues of the silicate magma (Papers III). The increase in the Si activity and decrease in the Al activity, by the mechanism referred to above, lead to the transition from calcic plagioclase to alkali feldspar and quartz crystallization, which dominated the crystallization history of the peralkaline granite magma (Papers II and III). As the magma was poor in Mg and rich in Fe and peralkaline, the mafic minerals were most likely Na- and Fe-rich variants of clinopyroxene (aegirine-augite–aegirine) and/or amphibole (riebeckite-arfvedsonite). Massive alkali feldspar dominant crystallization led to a sharp decrease in the Al, Na and K contents and overall peralkalinity and a relative increase in the Si, Ca, and Fe contents in the residual melts, resulting in bulk compositions approaching those of the Otanmäki mineralized rock units, although the observed high Ca enrichment likely required an additional mechanism (see below) (Paper III). The differences in the major element chemistry between the Kontioaho and Katajakangas mineralized rock units is attributed to the distinct degrees of differentiation attained in the residual melt before their separation and emplacement (Paper III). Such a FC model is also compatible with the observed volumes of the mineralized rocks, which are notably larger at Kontioaho compared to Katajakangas, being consistent with a FC-only process in which more highly evolved melts occupy smaller volumes (Paper III). This model implies that the two mineralized rock units likely represent fractionation products of separate batches of the parental peralkaline granite magma, which attained varying degrees of fractionation (Fig. 7b; Paper III).

The REE-HFSE mineralization in the Otanmäki system also appear to have required additional enrichment by fluoride, carbonate and sulphate complexing (Paper III), which has also been identified in many other studies of peralkaline REE-HFSE-rich granites and experiments as a process contributing to REE-HFSE mineralization and Ca enrichment (see Chapter 2.4). Differing compositions of the volatile phase in the Kontioaho (F-intensive) and Katajakangas (CO_2-S-intensive) occurrences also seem to explain their differences in Zr, Nb and REE enrichments (Paper III), which is supported by similar observations made in some experimental studies on silicate melts (e.g., Farges, 1996; Aseri et al., 2015).

The final step in the enrichment process was accumulation of the last residues of the REE-HFSE-rich silicate melt, apparently near the roof of the source peralkaline granite intrusions, followed by their emplacement (Paper III) (Fig. 7b). A similar setting is typical of REE-HFSE mineralization in many other REE-HFSE-rich peralkaline granite plutons where they occur in late aplite- and/or pegmatite-sheets above the intrusion roofs and along zones of structural weaknesses (e.g., faults) (e.g., Dostal et al., 2014; Vasyukova and Williams-Jones, 2014).

An enigmatic feature of the Otanmäki mineralization is the lack of evidence for significant REE-HFSE dispersion or metasomatic alteration effects even in the immediate wall rocks (Paper III). This is a rather striking feature considering the inferred mineralization model, which involves highly fractionated, felsic residual melts containing considerable amounts of dissolved volatiles (Paper III). Such melts can exsolve fluids capable of leaching REE and HFSE from earlier formed, primary magmatic phases, to be precipitated in fractures and cavities (cf. Dostal et al., 2014; Kempe et al., 2015; Vasyukova and Williams-Jones, 2018). Because such features are not observed in the Otanmäki system, any magmatic fluids that may have exsolved during late stages of crystallization of the peralkaline granite intrusions appear to have been either volumetrically minor or uncapable of mobilizing REE-HFSE from their highly refractory primary (magmatic) carrier minerals (e.g., zircon, allanite-(Ce)) or both (Paper III).

The current model of REE-HFSE enrichment to a wt% level at Otanmäki is based on geological observations, whole-rock chemical, Sm-Nd isotope chemical and U-Pb zircon geochronology, but these data still constitute a rather limited base of information for understanding the magmatic evolution and REE-HFSE enrichment processes. Obtaining further relevant information is challenging mainly because of the poor exposure and limited diamond drilling coverage of the REE-HFSE mineralization and alleged parental peralkaline granite. These hindrances make geological mapping and sampling of the mineralization difficult. Furthermore, the geology of the mineralization is complicated by effects of Svecofennian faulting. The metamorphic overprinting limits the usefulness of such approaches as mineral chemical and melt and fluid inclusion studies, which have been successfully used in genetic studies of many other (unmetamorphosed, non-deformed) REE-HFSE-rich peralkaline granites (e.g., Kovalenko et al., 1995; Marks et al., 2004; Vasyukova and Williams-Jones, 2014, 2018; Siegel et al., 2017b, 2018).

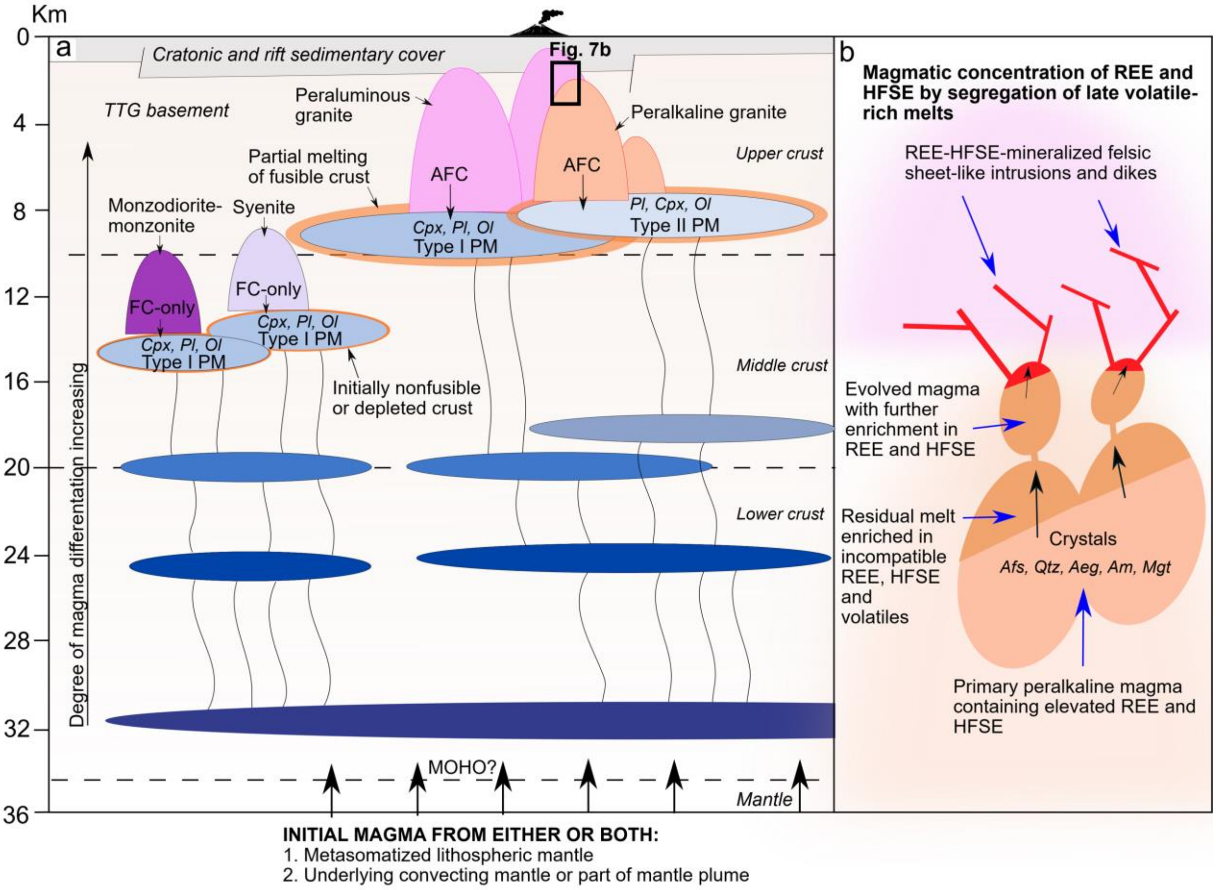

Figure 7. a) A schematic model of the petrogenesis of Otanmäki suite A1-type granites and associated intermediate rocks (monzodiorite-monzonite, syenite) by differentiation of mantle-derived parental magmas in the middle to upper crust (~2–4 kbar, ~7–15 km) (Paper II). The intermediate rocks were formed by fractional crystallization (FC) of type I parental magma involving crystallization of large amounts of clinopyroxene (Cpx), plagioclase (Pl) and olivine (Ol) with minimal crustal contamination (Paper II). The generation of the granites requires coupled fractional crystallization and assimilation of partial melts from fusible crust, such as Archean TTGs. The peraluminous granite is generated by assimilation fractional crystallization (AFC) from a relatively primitive parental magma (type I), which produces more clinopyroxene and olivine relative to plagioclase alongside with assimilation of high-silica partial melts from TTGs (Paper II). The genesis of the peralkaline granite requires a high degree of AFC of an already fractionated parental magma (type II), involving crystallization of a plagioclase-dominated mineral assemblage in the subsequent fractionation processes (Paper II). b) A schematic model of the origin of the Otanmäki REE-HFSE mineralization by extreme magmatic differentiation of a primary REE-HFSE- and volatile-rich peralkaline granite magma by alkali feldspar (Afs)-dominant crystallization within separate batches of the parental peralkaline granite magma, which underwent varying degrees of fractionation (Paper III). The final step in the enrichment process was accumulation of the last residues of the REE-HFSE-rich silicate melt, apparently near the roof of the source peralkaline granite intrusions, followed by their emplacement as a REE-HFSE-mineralized felsic sheet-like intrusions and narrow dikes above the intrusion roofs (Paper III). Qtz = quartz, Aeg = aegirine, Am = amphibole, Mgt = magnetite.

7.4 Implication for exploration

Economic aspects of this thesis project are related to the REE-HFSE potential of the peralkaline granites in the Otanmäki suite, such as those parental to the REE-HFSE mineralized rock units at Kontioaho and Katajakangas (Paper III). However, the magmatic evolution, which culminated in the generation of the Otanmäki REE-HFSE mineralization, appear to have been unfavorably different from the ideal "recipe" for creating a potentially mineable peralkaline granite-related REE-HFSE occurrence (see Chapter 2.4 and Vasyukova and Williams Jones, 2020) and therefore seems to have resulted in a seemingly lower degree of REE-HFSE enrichment in the Otanmäki system (Paper III) compared to that in some other peralkaline REE-HFSE-rich granites. In particular, the Otanmäki peralkaline granite and the REE-HFSE mineralized rock units lack the extreme magmatic Na-enrichment and late-magmatic hydrothermal dispersion and reprecipitation of REE-HFSE, which are shown in other studies to have significantly contributed to the size and grade of the resources, in some cases lifting them up to potentially mineable levels (e.g., Estrade et al., 2014; Gysi et al., 2016). In some cases, late-magmatic hydrothermal processes also favorably altered the magmatic REE-HFSE minerals to secondary minerals better suited to current beneficiation and hydrometallurgical processing, which require the rare earth elements to be preferably hosted either by REE fluorocarbonates (e.g., bastnäsite) and/or phosphates (e.g., monazite, xenotime) (Weng et al., 2015; Peiravi et al., 2021).

Although the Otanmäki system seems to be currently a less than "ideal" REE-HFSE exploration target (Paper III), it does not mean that the system could not host potentially minable REE-HFSE deposits. The system still remains underexplored for REE-HFSE and the obtained results in Papers I-III are useful in directing future exploration of REE-HFSE resources in the Otanmäki area. Both at Kontioaho and Katajakangas, one major problem concerning utilization is that the rare earth elements are hosted mainly by allanite-(Ce), which has not yet been exploited anywhere for the REE in an industrial scale as it cannot be economically processed by the presently established REE extraction methods (e.g., Verbaan et al., 2015; Demol et al., 2019; Peiravi et al., 2021). This means that turning the Otanmäki REE-HFSE occurrences into real economic resources would require finding a practical way to recover rare earth elements from allanite, and realistically taken, also an increase in the price of REEs. If such a technical and economic development was realized, the status of the Otanmäki area as an exploration target would significantly improve, especially noting the historical and recent discoveries of REE-HFSE-mineralized glacial boulders in the area (Repo, 2020), which suggest that there is a good potential for finding new occurrences of allanite-type REE mineralization in the Otanmäki A-type granites.

8 Conclusions

Based on the information presented in this dissertation, the following main conclusions can be drawn:

1) The Otanmäki suite comprises a broad variety of gneissic-textured intermediate and felsic igneous rocks including monzodiorite-monzonite, syenite and peralkaline to peraluminous granites, not just ‘alkaline granite’ as was previously thought. These igneous rocks display geochemical characteristics of ferroan A1-type granites typically generated in continental rift settings by differentiation of oceanic island basalt-type magmas with variable crustal contamination. The original igneous suite was metamorphosed under amphibolite facies conditions in Svecofennian tectono-thermal events and tectonically dismembered into two fault-bound units (nappes), which are now sliced between Archean TTG complexes and Paleoproterozoic supracrustal units of the Kainuu belt.

2) New geochronological investigations have clarified the age of the Otanmäki suite magmatism, showing that it took place at ca. 2.04–2.06 Ga. It records a rare event of intermediate to felsic non-orogenic plutonic magmatism; rocks of similar composition and age seem to be missing in other parts of the western margin of the Karelia craton and also in the Fennoscandian Shield as a whole. The general rarity of the Otanmäki suite igneous rocks is explained by the interpretation that the nappes containing the A1-type rocks represent long-travelled thrust sheets from a more distal, now disappeared part of the western Karelia margin.

3) Based on geochemical and thermodynamic modeling, the origin of the Otanmäki suite igneous rocks is explained by differentiation of oceanic island basalt-like mantle-derived parental magmas by fractional crystallization coupled with assimilation of Archean wall rocks, resulting in a spectrum of rocks representing residual melts or their mixtures with cumulate minerals. The outcomes of the FC-AFC modeling are consistent with previous studies suggesting that magmas of A-type ferroan granites become more peraluminous by involvement of partial melts of quartzo-feldspathic crust, such as Archean TTGs. The modeling also suggests that the differences in the parental melt

composition, which affect the quantity of late plagioclase fractionation, can explain the variation in the alkalinity and aluminosity of the end-product ferroan A-type granites, even in cases where there was a similar degree of crustal assimilation.

4) The Otanmäki suite granites host two targets of rare earth element (REE) and high-field strength element (HFSE) mineralization, localized in highly fractionated felsic dikes (Katajakangas) and a sheet-like intrusion (Kontioaho) containing allanite-(Ce)-zircon-titanite-Nb-REE-Th-U oxide assemblages. Allanite-(Ce) is recognized as the major host for REEs in both occurrences. The new results reveal that the REE-HFSE enrichment was intrinsically related to the peralkaline magmatism of the Otanmäki suite and represents highly evolved fractionation products from a parental peralkaline felsic magma that was enriched in REE-HFSE and volatile elements (e.g., F, CO_2, S).

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Appendix

List of diamond drill hole locations.

HOLE ID	LENGTH (m)	X_KKJ	Y_KKJ	YEAR	AZIMUTH	DIP
KATAJAKANGAS-001	**85.02**	3503230	7114095	1973	23	39
KATAJAKANGAS-002	**203.10**	3503385	7113463	1983	23	59
KATAJAKANGAS-003	**156.50**	3503529	7113428	1983	23	68
KATAJAKANGAS-004	**210.30**	3501300	7114250	1983	360	67
KATAJAKANGAS-005	**195.20**	3501300	7113988	1983	360	68
KATAJAKANGAS-006	**178.90**	3501300	7114530	1984	360	68
KATAJAKANGAS-007	**179.00**	3500939	7113967	1984	335	59
KATAJAKANGAS-008	**177.30**	3501085	7113661	1984	335	59
KATAJAKANGAS-009	**203.10**	3502636	7113575	1984	22	64
KATAJAKANGAS-010	**202.60**	3501825	7113440	1984	360	50
KATAJAKANGAS-011	**103.80**	3501300	7114110	1984	360	64
KATAJAKANGAS-012	**55.40**	3501200	7114198	1984	360	61
KATAJAKANGAS-013	**134.60**	3501200	7114019	1984	360	63
KATAJAKANGAS-014	**68.70**	3501100	7114110	1984	360	63
KATAJAKANGAS-015	**126.00**	3501100	7113998	1984	360	63
KATAJAKANGAS-016	**80.70**	3500854	7113927	1984	335	56
KATAJAKANGAS-017	**107.80**	3500899	7113829	1984	336	54
KATAJAKANGAS-018	**138.40**	3500905	7113816	1984	336	54
KATAJAKANGAS-019	**110.20**	3501400	7114199	1985	360	63
KATAJAKANGAS-020	**155.20**	3501400	7114080	1985	360	63
KATAJAKANGAS-021	**122.40**	3501500	7114200	1984	360	63
KATAJAKANGAS-022	**188.80**	3501500	7114070	1985	360	64
KATAJAKANGAS-023	**335.60**	3501400	7114800	1985	360	58
KONTIOAHO-001	**137.45**	3500380	7116350	1980	306	54
KONTIOAHO-002	**165.40**	3500050	7117650	1980	306	55
KONTIOAHO-003	**210.70**	3501829	7114987	1984	45	70
KONTIOAHO-004	**144.30**	3502038	7115195	1984	45	64
KONTIOAHO-005	**159.00**	3501350	7116075	1984	52	63
KONTIOAHO-006	**148.00**	3499745	7115720	1984	297	51
KONTIOAHO-007	**184.60**	3500219	7116427	1984	305	56
KONTIOAHO-008	**199.30**	3501010	7116408	1984	51	53
KONTIOAHO-009	**43.50**	3502186	7115342	1984	45	63
KONTIOAHO-010	**30.90**	3502236	7115306	1984	45	63
KONTIOAHO-011	**110.70**	3502150	7115169	1984	45	63
KONTIOAHO-012	**131.50**	3501208	7115964	1985	52	54
KONTIOAHO-013	**150.50**	3501544	7115720	1985	45	63
KONTIOAHO-014	**233.70**	3500420	7115680	1984	306	72
JOKIKANGAS-001	**148.80**	3499058	7114483	1984	305	53
JOKIKANGAS-002	**126.50**	3499446	7114195	1984	305	53
JOKIKANGAS-003	**158.30**	3499260	7114324	1984	305	50
JOKIKANGAS-004	**190.30**	3500387	7113569	1984	305	51

JOKIKANGAS-005	**194.20**	3500630	7113770	1984	307	53
JOKIKANGAS-006	**228.60**	3499130	7113550	1984	306	47
JOKIKANGAS-007	**157.70**	3498560	7112958	1984	317	63
JOKIKANGAS-008	**151.40**	3497903	7113900	1984	90	47
JOKIKANGAS-009	**57.50**	3497720	7113900	1984	90	49
JOKIKANGAS-010	**188.70**	3497315	7114160	1984	54	45
JOKIKANGAS-011	**182.00**	3496595	7113800	1984	90	41
JOKIKANGAS-012	**128.00**	3496755	7113815	1984	90	45
JOKIKANGAS-013	**177.00**	3496826	7114194	1984	90	45
JOKIKANGAS-014	**124.00**	3499242	7113467	1984	306	54
HONKAMÄKI-R20	**209.60**	3495560	7116154	1978	297	54
HONKAMÄKI-R21	**90.40**	3495581	7116303	1978	297	54
HONKAMÄKI-R22	**98.3**	3495655	7116370	1978	297	54
HONKAMÄKI-R23	**106.6**	3495708	7116456	1978	297	54
HONKAMÄKI-R24	**129.3**	3495512	7116050	1978	297	54
HONKAMÄKI-R25	**153.8**	3495433	7115896	1978	297	54
HONKAMÄKI-R26	**250.20**	3496111	7115850	1978	297	54
HONKAMÄKI-R27	**110.7**	3495396	7115791	1978	297	54
HONKAMÄKI-R28	**108.7**	3495320	7115716	1979	297	45
HONKAMÄKI-R29	**168.30**	3496177	7115812	1979	297	54
HONKAMÄKI-R30	**101.30**	3495887	7115857	1979	297	45
HONKAMÄKI-R31	**145.30**	3496212	7116127	1979	297	50
HONKAMÄKI-R32	**121.20**	3496001	7116355	1979	298	50
HONKAMÄKI-R33	**144.00**	3495782	7114462	1979	298	50
HONKAMÄKI-R34	**136.35**	3495858	7114421	1979	298	50
HONKAMÄKI-R35	**155.80**	3496160	7115487	1985	297	45
HONKAMÄKI-R36	**154.50**	3496252	7115435	1985	297	45
HONKAMÄKI-R37	**110.70**	3496310	7115407	1985	297	44
HONKAMÄKI-R38	**174.60**	3496454	7115341	1985	297	46
HONKAMÄKI-R39	**150.30**	3496329	7116071	1985	297	45
ETELÄSUO-001	**178.1**	3507951	7113880	1979	270	63
ETELÄSUO-002	**121.5**	3508000	7113780	1979	270	63
ETELÄSUO-003	**165.40**	3509350	7115300	1985	180	48
ETELÄSUO-004	**81.10**	3508591	7114964	1985	180	45
ETELÄSUO-005	**20.30**	3508600	7115220	1985	243	45
ETELÄSUO-006	**160.00**	3508476	7115157	1985	63	45
ETELÄSUO-007	**150.70**	3508370	7115103	1985	63	48
ETELÄSUO-008	**155.20**	3508267	7115050	1985	63	45
PAPUSUO-R1	**180.00**	3496229	7112797	1977	135	45
PAPUSUO-R2A	**196.60**	3497727	7113230	1977	135	45
PAPUSUO-R3	**124.80**	3497565	7113527	1977	135	45
SAUNAKANGAS-001	**123.50**	3506000	7117100	1979	360	54
VIMPELINJOKI_R-001	**394.6**	3507425	7115404	1979	270	63
VIMPELINJOKI_R-002	**127.3**	3506811	7115399	1979	270	63
ISONKIVENKANGAS_R-001	**161.4**	3496620	7119360	1978	270	45

ISONKIVENKANGAS_R-002	**70.8**	3496590	7119000	1980	270	50
PENTINPURO-R1		3497327	7121494			
PENTINPURO-R2		3497230	7121431			
PENTINPURO-R3	**81**	3497103	7121269	1945	296	45
PENTINPURO-R4	**91.15**	3497146	7121359	1945	296	49
PENTINPURO-R5	**109.3**	3497885	7115100	1980	291	50
ITÄRANTA-R1	**214.6**	3516578	7113415	1978	50.4	54
ITÄRANTA-R2		3518750	7110806			
ITÄRANTA-R3	**59.6**	3519580	7110283		23	45
ITÄRANTA-R4	**99.4**	3519567	7110539			45
ITÄRANTA-R5	**105.8**	3519828	7110683		140	54
ITÄRANTA-R6	**115.5**	3519692	7110848		176	45
ITÄRANTA-R7	**85**	3519512	7110550		140	45
ITÄRANTA-R8	**80.1**	3519637	7110531		140	45
ITÄRANTA-R9	**52.3**	3519684	7110576		140	45
ITÄRANTA-R10	**186.75**	3519596	7110495		140	45
ITÄRANTA-R11	**202.95**	3516500	7113488	1978	50.4	54
ITÄRANTA-R12	**173.3**	3516683	7113400	1978	50.4	54
ITÄRANTA-R13	**156.35**	3515987	7114499	1978	50.4	54
VUOROKAS-R97	**115.9**	3508298	7112272		140	54

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