Contributions to Mineralogy and Petrology

A Nd- and O-isotope study of the REE-rich peralkaline Strange Lake granite: Implications for Mesoproterozoic A-type magmatism in the Core Zone (NE-Canada) --Manuscript Draft--

Manuscript Number:	CTMP-D-17-00008R1	
Full Title:	A Nd- and O-isotope study of the REE-rich peralkaline Strange Lake granite: Implications for Mesoproterozoic A-type magmatism in the Core Zone (NE-Canada)	
Article Type:	Original Paper	
Keywords:	Nd isotopes, O isotopes, Strange Lake, REE, Core Zone, Nain Plutonic Suite	
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Funding Information:	Canadian Network for Research and Innovation in Machining Technology, Natural Sciences and Engineering Research Council of Canada	Prof. Anthony E. Williams-Jones
Abstract:	It is well established that A-type granites enriched in high field strength elements, such as Zr, Nb and the REE, form in anorogenic tectonic settings. The sources of these elements and the processes controlling their unusual enrichment, however, are still debated. They are addressed here using neodymium and oxygen isotope analyses of samples from the 1.24 Ga Strange Lake pluton in the Paleoproterozoic Core Zone of Québec-Labrador, an A-type granitic body characterised by hyper-enrichment in the REE, Zr, and Nb. Age-corrected ϵ Nd values for bulk rock samples and sodic amphiboles (mainly arfvedsonite) from the pluton range from -0.6 to -5.7, and -0.3 to -5.3, respectively. The ϵ Nd values for the Napeu Kainiut quartz monzonite, which hosts the pluton, range from -4.8 to -8.1. The 147Sm/144Nd ratios of the suite and the host quartz monzonite range from 0.0967 to 0.1659, large variations that can be explained by in-situ fractionation of early LREE-minerals (Strange Lake), and late hydrothermal HREE remobilization. Oxygen isotope analyses of quartz of both Strange Lake and the host yielded δ 180 values between +8.2 and +9.1, which are considerably higher than the wantle value of 5.7 ± 0.2 ‰. Bulk rock oxygen isotope analyses of biotite-gneisses in the vicinity of the Strange Lake pluton yielded δ 180 values and positive δ 180 values of the Strange Lake and Napeu Kainiut samples indicate that both magmas experienced considerable crustal contaminants were sedimentary-derived rocks from the underlying Archean Mistinibi (para-) gneiss complex, which is characterized by low ϵ Nd and high δ 180 values. Mixing of 5 - 15 % of a gneiss, having an ϵ Nd value of -15 and $a \delta$ 180 values between +11, with a moderately enriched mantle source (ϵ Nd = +0.9, δ 180 = +6.3) would produce values similar to those obtained for the Strange Lake pluton and associated REF-	

	mineralized anorogenic bodies formed from a combination of subduction-induced fertilization of the sublithospheric mantle, crustal extension and in-situ magma evolution.
Response to Reviewers:	RESPONSE TO REVIEWERS We are encouraged that Editor Timothy L. Grove and Reviewer #1 consider our manuscript, which presents a hypothesis for the generation of the Strange Lake pluton and other HFSE mineralized silicate bodies of the Nain Plutonic Suite, to have merit and be worthy of publication after appropriate revision. Reviewer #2 questions our model that mantle-derived fertile melts were important and requests additional proof. Below, we systematically address the minor points raised by Reviewer #1 and then the more substantial criticisms of the manuscript by Reviewer #2.
	REVIEWER # 1 General comments/questions of Reviewer # 1: Reviewer #1 considers our study, the data and the illustrations to be of very good quality. He finds our arguments convincing, and commends us for our awareness that our model may not be the only model that could explain the generation of the Strange Lake magma. His only concerns relate to the illustrations. These concerns are addressed below. Minor comments: Lines 268-269 vs. Figure 3. The figure should be corrected, as quartz monzonite samples from outside the pluton are shown as having higher REE contents than in the roof pendants inside the pluton, contrary to what is listed in Table 1 and written (correctly) in the text.
	The colours for the quartz monzonite in Figure 3 have been corrected.
	Figures 3-6. Colours used for hypersolvus and transsolvus granite samples are too similar and cannot be discriminated easily, especially if the figures suffer reduction in the published article. The colour of the transsolvus granite samples in Figures 3-6 has been enhanced in order to provide greater contrast.
	Figure 5. Quench zone samples shown in the figure are not listed in Table 2, except sample 10036-WR. Is it because their 147Sm/144Nd ratios are higher than 0.13 (cf. line 389)? The diagram in Figure 5 includes three quench zone samples from the dataset of Kerr (2015), which are not listed in Table 2. The colours of these samples in Figures 4 and 5 have been changed to light grey in order to identify them as being quench zone samples (the same colour was used previously in Fig. 3 for this purpose).
	REVIEWER # 2 The general comments/questions of reviewer # 2 are: Reviewer #2 is generally opposed to our hypothesis that the Strange Lake (SL) granite was derived largely from subcontinental lithospheric mantle (SCLM), which was initially depleted and subsequently enriched by metasomatic fluids produced by subduction- related melts. He favours a model in which Strange Lake was derived from (likely metasomatized) crustal melts and would like to see us consider the possibility that the Napeu Kainiut quartz monzonite (host rock) represents this melt. His issues with our manuscript ultimately stem back to the debate over whether A-type granites are derived solely from crustal melts or contain a significant mantle component (see introduction, lines 31-46).
	1.Reviewer #2 criticizes the lack of information on the major and trace element composition of the Strange Lake pluton and the Napeu Kainiut host rock. We accept the validity of this criticism and have added a table reporting the average major and trace element contents for each rock unit discussed in the manuscript.
	2.He disagrees with our description of the REE profiles for the Strange Lake pluton as being flat (implying a mantle-like pattern). We agree and have changed the text to provide a more accurate description of the profile. The reviewer points out that due to 'elevated LREE and flat HREE, with a pronounced Eu anomaly', plagioclase is required in the source during partial melting and that the SL granite was thus produced from the crust and not the mantle. We understand that melting of a source with residual

plagioclase would produce a liquid with a negative Eu-anomaly. However, considering the broader intrusive history of the Nain Plutonic Suite, which, among other things records the development of large anorthosite complexes, it seems simpler to interpret the negative Eu-anomaly as originating from fractional crystallization of large masses of plagioclase early in the history of the suite.

3. The reviewer states, that the strong similarity of the Napeu Kainiut REE profile to that for the Strange Lake pluton, albeit at lower absolute values, is not discussed. This is not true. The profiles of the two rock groups are compared and discussed on lines 256 - 262 and 405 - 408. Moreover, an entire section starting at line 398 and continuing to line 420 is devoted to a discussion of the relationship of the Strange Lake granites to the Napeu Kainiut host rock.

4. The reviewer states that a cursory examination of published data on Garder and Nain basaltic dykes by the reviewer revealed 'very different, moderately enriched, smooth REE patterns, but with much lower total REE abundances' to him. He argues that it is not obvious that the granite REE contents could have been derived from this magma. Indeed, the chondrite normalized REE pattern of the Nain and Gardar basaltic dikes are rather flat, but with slightly elevated LREE, and much lower total concentrations than the SL granites (e.g. 1.73 Harp dikes, Cadman et al., 1993, Fig. 7d and Wiebe, 1985, Fig. 4a; Gardar B0 dikes, Bartels et al., 2015). Our interpretation of the source of the Strange Lake magma is based on more than chondrite-normalized REE profiles. and contrary to the opinion of the reviewer, who argues for a crustal source for the melt, we do have evidence for a mantle influence. Indeed, our Nd-isotope dataset (e.g., initial ɛNd value slightly negative and close to zero) provides a clear indication that both mantle and crustal sources played an important role in the genesis of the Strange Lake granites. However, we do not rule out the possibility that a lower crustal melt fertilized by mantle fluids (e.g., Martin, 2006) also could have been the source of the Strange Lake magma (see lines 423, and 430, respectively). Finally, as the profiles of these mafic rocks are rather flat but slightly LREE enriched, they are not very different in form to those for the Strange Lake rocks, and therefore we disagree with the statement that the rocks are clearly not related.

5. The reviewer proposes that we consider the hypothesis that the Napeu Kainiut host, or its source, was the source of the Strange Lake magma based on a similar Sm/Nd ratio (Fig. 4) and δ 18O composition (~9 ‰). He proposes that we model the Napeu Kainiut host as the evolved end-member in Figs. 9a, b and Fig. 11. He states that this would remove the obvious problem of why none of the Strange Lake rocks have δ 180 values >9 %.. What is required is that such a source rock melt and mix with relatively primitive SCLM, which is what Fig 6 (a,b) shows. The contrast in Fig 5 simply shows similar fractionation slopes for SL and NK, but different parental magma compositions different proportions of endmember components.' The reviewer seems to have missed the very important point that the δ 18O values of the Napeu Kainiut 8.9 – 9.1 ‰ are within the range of the values for the Strange Lake granite 8.4 – 9.1 ‰. As the mantle value is considerably lower (6.1 ‰), it therefore follows that even minimal mixing between these two endmembers will produce a magma with a δ 180 value lower than that of the upper end of the range for Strange Lake rocks. Indeed, our modelling (Figure 9) shows convincingly that the crustal endmember would require a δ 180 value > 10 to explain the values for observed for Strange Lake, which effectively rules out the Napeu Kainiut as an endmember. This issue is illustrated and discussed in Fig. 9a and in lines 551 -563, respectively.

6.In this comment the reviewer states that irrespective of what model you use, neither adequately explains the REE enrichment in the SL pluton. The general model of refertilised SCLM suffers from the fact that many post-collisional granites are postulated to have been derived from such a source, but few have the extreme REE values described here. A long-lived "refertilised" source from prior subduction should also be hydrated, unless special pleading is introduced. It was not the purpose of our study to explain the extreme REE enrichment in the Strange Lake pluton. However, we disagree that neither model can explain such extreme REE enrichment. Indeed, we consider it very likely that a very small degree of partial melting of a metasomatised (F-enriched) mantle in combination with extreme fractional crystallisation (see Boily and Williams-Jones, 1994) could very well explain this enrichment. The same could be said for a crustal source adequately fertilised by mantle fluids.

7.The reviewer states that the other glaring problem is that the authors argue for low degrees of partial melting yet the early phase granite is hypersolvus, whereas it should be a volatile-rich eutectic melt. Therefore the reasons for the REE enrichment in the pluton remain enigmatic. Indeed, we do argue for a low degree of melting, however, we also propose that a significant proportion of silicic crust was added to the initial mantle melt, which subsequently underwent fractionation and produced the highly evolved Strange Lake pluton. There is thus no reason to assume that it was an eutectic melt. The hypersolvus granite at SL does, in fact, exhibit elevated F, OH and other volatiles, which are evident in abundant fluorite, arfvedsonite (OH) and from fluid inclusion studies (CH4). The water fugacity, however, was obviously too low to cause crystallization of two alkali feldspars. The question of why the pluton is so enriched in the REE has been addressed in a number of studies (e.g., Boily and Williams-Jones, 1994), and most recently, by Vasyukova and Williams-Jones (2014, 2016) in which a case is made for the role in this of silicate-fluoride melt immiscibility.

Specific comments:

Line 413. You say "the model ages reveal that initial melt extraction from the mantle was much later for the Strange Lake granites that for the rocks of the Napeu Kainiut pluton", based on Fig. 7. I look at that and consider that the variable ϵ Nd values for the SL pluton suggests mixing, as shown in Fig. 6, and that a higher proportion of mantle is involved in SL versus NK. However, both could have comes from similar endmember components. Hf isotopes in zircon would extend the array and probably solve this issue. Thus, I don't see why initial melt extraction from the mantle had to be earlier in NK. Not sure what you are trying to say.

Reviewer 2 argues that the range in model ages for the SL granites and the Napeu Kainiut pluton reflects different mantle crust mixtures involving a crustal magma (Napeu Kainiut) and mantle magma. We considered this possibility in lines 415-419 and have slightly modified the statements slightly so that the point is made more clearly. However, we stress that field relationships indicate that the SL granites are younger than the Napeu Kainiut and the geochemistry of the Napeu Kainiut is consistent with the older (1.42 Ga) Mistastin intrusions.

Line 420. Actually, you do suggest that both NK and SL could be derived from a similar source, but if they came from the same source, then plagioclase was present in the source, requiring a dominantly crustal endmember. But why did the SL pluton became systematically more enriched in the REE?

Reviewer 2 seems to argue for crustal melting with residual plagioclase in order to produce the negative Eu anomalies. Given that numerous studies have demonstrated a high degree of fractional crystallization within the SL granites, an origin of the negative Eu anomalies by plagioclase fractionation seems simpler and more straightforward. The goal of the paper is to discuss the relative contributions of mantle and crust to the SL pluton. The REE enrichment processes of the Strange Lake pluton have been discussed in Gysi and Williams-Jones (2013); Salvi and Williams-Jones (1990); Salvi and Williams-Jones (1996); Vasyukova and Williams-Jones (2014) amongst others.

Line 421. You say "The extreme REE, Zr, Nb and alkali enrichment of the Strange Lake pluton precludes its origin solely by melting of crustal material". I don't understand this statement. Are you saying that crustally-derived magmas cannot be so REE rich? Why? I would have thought the ϵ Nd isotopic array provided more definitive evidence for this interpretation (ie., a crust-mantle mix).

Indeed, it is theoretically possible to produce elevated HFSE and alkalis from the melting of an entirely crustal source as concluded by some researchers (see introduction, lines 39-42). However, as pointed out in this manuscript, our Nd isotope dataset provides clear evidence for a crust-mantle mix. We have rewritten the sentence to clarify this point.

Line 441. You say "Most of the intrusions of the Nain Plutonic Suite, including the Strange Lake granites, have relatively flat chondrite-normalized REE profiles (Fig. 3)." Well, this is wrong, and defies your earlier description. Even the mafic Gardar dykes are described thus: "Chondrite-normalized REE plots display a significant enrichment of REE which is more pronounced for the light rare-earth elements (LREE) with La abundances between 11-16 times chondritic values" (Bartels et al). This REE pattern is nothing like the SL pluton (much lower contents), but is typical of continental tholeiites. The SL granites do not have a mantle signature, based on the REE patterns. See comment above.

We accept the criticism and in the revised manuscript provide more accurate descriptions of the REE profiles.

Line 454 You go on to say "The REE profiles for the Strange Lake granites, are thus consistent with the melting of garnet-poor lherzolite or garnet-free lherzolite at depths less than those for which garnet is stable." Do you really believe that the SL pluton is derived from Iherzolite??!! This is what happens when no major element chemistry is included to provide mass balance considerations. I believe this is the case for the mafic rocks of the Nain suite, but to extend it to the granites is beyond logic. The large Eu anomaly demands that plagioclase be a persistent and extensive component during melting of the source. Line 441 drives you toward false assumptions. Our intention was to point out that the SL granite formed via fractionation of a mafic mantle derived melt with accompanying crustal contamination. We have modified the phrase accordingly. As mentioned earlier, we consider it more likely that the strong negative Eu anomaly is evidence of plagioclase fractionation, not source. Line 625. As part of the tectonic model, the sequence includes: "1) subduction of basaltic (alkali- and volatile-rich) oceanic crust and 2) fertilization of subcontinental mantle with alkalis and volatiles (F, Cl, H2O, CO2). First of all, what is alkali and volatile rich oceanic crust? Where does it exist? Most oceanic crust is MORB, not alkali-rich. Alkali-rich basalts are typically OIBs, which form at hotspots and along leaky transforms; they are not fluid-rich in general, nor are they abundant, so there is a conundrum. Also, the fluids in subduction zones are typically H2O rich, not just halogens, so how do you concentrate the halogens and remove the water? Moreover, the hypothesised subduction-related scenario is not different to any post-collisional setting, where granites can be S- (peraluminous) or I-type (metaluminous); rarely peralkaline.

Altered seafloor basalt is enriched in alkali elements (K, Na) and volatiles (H2O, Cl), and is represented in predominantly basaltic ocean floor with little sediment cover. Etype (enriched) MORB is enriched in K and the HFSE (Sun and McDonough 1989). Alkali and HFSE enrichment of MORB-type oceanic crust prior to subduction could have occurred by mantle upwelling and the intrusion of non-depleted magmas from lower mantle regions. Moreover, it is not necessary for the subducted ocean crust to be enriched in CO2 or be an alkali basalt. This can all be generated in the extensional phase of the model. Alkali rich basalts are not ubiquitously of OIB origin, as the example of the Eriksfijord basalts in the Gardar Province shows (Halama et al. 2003). The trace element profiles of these basalts are intermediate between E-type MORBs and OIBs and might well represent a mixture of depleted and enriched mantle. This is also evident in the Sr and Nd isotope signatures of these rocks.

Elevated F contents have been documented in subduction-related mafic dykes of the Gardar province. There it was concluded that F was retained mainly in phlogopite in the source, which tends to concentrate F at the expense of OH (Köhler et al. 2009). The more recent subduction-related magmas of the Andes show comparably elevated F contents (e.g., Morgan et al., 1998). Thus F-enrichment may be typical for subduction related mantle-metasomatism.

In our model, we do not propose metasomatism of the SCLM to be the only cause of the resulting alkaline magmatism in the Nain Province. On the contrary, we emphasize the importance of a low degree of partial melting due to thickened lithosphere during continent-continent collision (lines 637 to 640), as well as on repeated post-collisional crustal extension. The latter represents the typical tectonic setting of alkaline (and peralkaline) magmas. We used the analogue of the Gardar Province, due to its proximity and the many similarities with the Nain Plutonic Suite for our model, where it has been shown repeatedly that the alkaline magmatism was affected by subduction-related metasomatism.

Line 640. What is the significance of this statement "This thickening of the crust exerted significant pressure on the underlying lithospheric mantle."

Rephrased. The statement is significant in as much as the enhanced pressure on the underlying mantle resulted in a lower degree of partial melting.

Line 652. What is syn-collisional crustal extension? Nonetheless, it is clear that the SL pluton and similar bodies of the Nain Suite are related to protracted crustal extension, and a distal backarc setting seems likely.

Rephrased. The wording 'syn-collisional' in this context was meant in terms of the magmatism that occurred during the Elsonian event, and not in respect to crustal extension.

Line 665. Reference to Andean subduction. There is not one reference to peralkaline granites in the Andes in the Mamani et al paper. I have not heard of them either. The

Permo-Triassic Choi-Choi province are the only examples I'm aware of there, but that is pre-Andean.

The point of the reference was not to show an association between alkaline granites and subduction, but to merely point out that subduction related magmas and associated mantle enrichment can reach as far as 400 km in-board of the subduction zone. There does not have to be an association with alkaline granites. This step is merely an enrichment mechanism.

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A Nd- and O-isotope study of the REE-rich peralkaline Strange Lake granite: Implications for Mesoproterozoic A-type magmatism in the Core Zone (NE-Canada)

Contributions to Mineralogy and Petrology

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ACKNOWLEDGEMENTS

Financial support for the research was provided by a grant from Quest Rare Minerals Ltd. and a matching grant from the NSERC Collaborative Research and Development program. Quest Rare Minerals Ltd. also provided essential logistical support in the field. André Poirier and Julien Gogot helped with the Nd isotope analyses, which were carried out in the GEOTOP laboratories (Université de Québec à Montréal). Galen Halverson (Earth & Planetary Sciences, McGill University) made his equipment in the clean laboratories available to the project and Peter Crockford helped with the wet column chemistry and the preparation of samples for the TIMS analyses (Sm-Nd-isotope analyses). Mike Hamilton (University of Toronto), Andy Kerr (formerly of the Newfoundland and Labrador Geological Survey), David Corrigan (Geological Survey of Canada) and Robert F. Martin (McGill University) contributed from their extensive knowledge and experience with alkaline igneous rocks, and provided supplementary data (unpublished maps and literature).

ABSTRACT

It is well established that A-type granites enriched in high field strength elements, such as Zr, Nb and the REE, form in anorogenic tectonic settings. The sources of these elements and the processes controlling their unusual enrichment, however, are still debated. They are addressed here using neodymium and oxygen isotope analyses of samples from the 1.24 Ga Strange Lake pluton in the Paleoproterozoic Core Zone of Québec-Labrador, an A-type granitic body characterised by hyper-enrichment in the REE, Zr, and Nb. Age-corrected ɛNd values for bulk rock samples and sodic amphiboles (mainly arfvedsonite) from the pluton range from -0.6 to -5.7, and -0.3 to -5.3, respectively. The ENd values for the Napeu Kainiut quartz monzonite, which hosts the pluton, range from -4.8 to -8.1. The ¹⁴⁷Sm/¹⁴⁴Nd ratios of the suite and the host quartz monzonite range from 0.0967 to 0.1659, large variations that can be explained by in-situ fractionation of early LREE-minerals (Strange Lake), and late hydrothermal HREE remobilization. Oxygen isotope analyses of quartz of both Strange Lake and the host yielded δ^{18} O values between +8.2 and +9.1, which are considerably higher than the mantle value of 5.7 ± 0.2 %. Bulk rock oxygen isotope analyses of biotite-gneisses in the vicinity of the Strange Lake pluton yielded δ^{18} O values of 6.3, 8.6 and 9.6 %. The negative ϵ Nd values and positive δ^{18} O values of the Strange Lake and Napeu Kainiut samples indicate that both magmas experienced considerable crustal contamination. The extent of this contamination was estimated, assuming that the contaminants were sedimentary-derived rocks from the underlying Archean Mistinibi (para-) gneiss complex, which is characterized by low ϵ Nd and high δ^{18} O values. Mixing of 5 – 15 % of a gneiss, having an ϵ Nd value of -15 and a δ^{18} O value of +11, with a moderately enriched mantle source ($\epsilon Nd = +0.9$, $\delta^{18}O = +6.3$) would produce values similar to those obtained for the Strange Lake granites. Based on analogies between the Nain Plutonic Suite and the Gardar alkaline igneous province (SW-Greenland), we conclude that the Strange Lake pluton and associated REE-mineralized

anorogenic bodies formed from a combination of subduction-induced fertilization of thesublithospheric mantle, crustal extension and in-situ magma evolution.

28 KEYWORDS

Nd isotopes, O isotopes, Strange Lake, REE, Core Zone, Nain Plutonic Suite

30 INTRODUCTION

Although it is well known that A-type granitic rocks are enriched in incompatible and high field strength elements (HFSE), such as the rare earth elements (REE), Zr, Nb, U, Ta and volatiles such as F and Cl (Eby 1990; 1992; Bonin 2007), the reasons for this enrichment continue to be a matter of considerable debate. Some researchers, on the basis of Pb, Nd and Sr isotopic evidence and trace element modeling, have concluded that A-type granites are produced by the fractionation of basaltic magmas of mantle origin (Frost et al. 1997; 1999; 2001; Anderson et al. 2003). Other researchers have interpreted them to result from melting of lower crustal material that has been metasomatized by fertile mantle-derived fluids (Woolley 1987; Martin 2006; 2012) . A few studies, noting the variability of initial ⁸⁷Sr/⁸⁶Sr ratios and other trace element variations, have proposed that A-type granites are exclusively of crustal origin (Collins et al. 1982; Dall'Agnol et al. 2005; Dall'Agnol and de Oliveira 2007). Finally, a considerable number of studies have argued that A-type granites crystallize from fertile mantle melts that have undergone variable degrees of crustal assimilation (Kovalenko et al. 2004; 2007; 2009; Hegner et al. 2010). According to this interpretation, an initially depleted mantle is metasomatically enriched in the REEs and alkalis as well as in volatiles such as H₂O, CO₂ CH₄ and F (Bailey 1987).

Most A-type granitic intrusions are emplaced in anorogenic intra-plate rift-settings, e.g., settings of crustal de-stressing following collisional events. For example, the large AMCG (Anorthosite-Mangerite-Charnockite-Granite) intrusions of the Nain Plutonic Suite (NPS) in Labrador and northern Québec were emplaced during the waning stages of a series of orogenies, i.e., the Pinwarian (1.52-1.46 Ga), the Elsonian (1.46-1.23 Ga) and the Grenvillian (1.08-0.99 Ga) orogenies (Gower and Krogh 2002; McLelland et al. 2010). Some researchers have proposed that the emplacement of anorogenic intrusive suites is the result of a combination of tectonic processes, e.g., continent-continent collision followed by the

This study investigates the genesis of A-type granites using the Strange Lake pluton in

delamination of a thickened lithosphere, the ascent of asthenosphere and subsequent rifting (Connelly and Ryan 1999; Bonin 2007; McLelland et al. 2010).

northern Québec-Labrador, which is composed of highly evolved peralkaline granitic intrusions, and late pegmatites that host two potentially economic REE-Zr-Nb deposits. One of the deposits, the B-zone, which is currently being evaluated for exploitation, is estimated to contain a resource of 278 Mt of ore, grading 0.93 wt. % REE₂O₃ (39% heavy rare-earth oxides), 1.92 wt. % ZrO₂ and 0.18 wt. % Nb₂O₅ (Quest 2012). The Strange Lake pluton is the youngest representative of the alkaline Nain Plutonic Suite, which was emplaced in Québec and Labrador between 1.46 and 1.24 Ga (Miller et al. 1997; Gower and Krogh 2002). Based on the data presented in this paper and the generally accepted plate tectonic framework for Mesoproterozoic alkaline magmatism in northeastern Laurentia, we have developed a model for the formation of the hyper REE- and HFSE-mineralized Strange Lake pluton and, by extension, for other alkaline plutons in the Nain and Gardar (a postulated equivalent of the Nain plutonic province in southern Greenland) igneous provinces.

Using a set of radiogenic neodymium, and stable oxygen isotope data for the unaltered Strange Lake granites from the Québec side (this study) and additional isotopic data from the Labrador side of the pluton (Kerr 2015), we show that the corresponding magmas originated from partial melting of a fertile mantle and evolved by the assimilation of continental crust. A combination of low degrees of partial melting of an enriched mantle, fractional crystallization and assimilation of underlying Archean gneiss satisfactorily explain the hyper-enrichment in the REE and the isotope signatures that we have obtained. In addition, published Nd and O isotope data and interpretations of the Nain Plutonic Suite and the Gardar alkaline province are consistent with our results, namely that the isotopic signature of the intrusions was greatly

affected by the age (Archean vs. Paleoproterozoic) and the nature (metasedimentary) of the underlying crust.

GEOLOGIC SETTING

82 Regional geology

As noted above, the Strange Lake pluton is part of the Nain Plutonic Suite, a large igneous province in northeastern Canada (Fig. 1). This province comprises anorthositic-mangeritic-charnockitic-granitic intrusive rocks (AMCG complexes) and straddles the boundary between Archean rocks of the Nain Province to the east and late Archean and Early Paleoproterozoic rocks of the southeastern Churchill Province to the west; the latter has more recently been referred to as the 'Core Zone' (Emslie et al. 1994; James et al. 1996). Beginning with the Michikamau and Harp Lake anorthosites and the Mistastin granitoid rocks (1.46-1.42 Ga), large batholiths were emplaced over a time span of about 220 Ma (Gower and Krogh 2002). These early intrusions were followed by the emplacement of anorthosites, troctolites, norites, diorites and granitoids (1.35-1.29 Ga) to the east. They, in turn, were crosscut by alkaline and peralkaline volcanics and plutons, e.g., Flowers River (1.27 Ga), and the Strange Lake pluton (1.24 Ga) (Fig. 1). The anorthosites, troctolites and norites were also intruded by small masses of ferrodiorite, which are interpreted to have formed from the residual liquids of anorthosite crystallization (Emslie et al. 1994; McLelland et al. 2010).

97 The Strange Lake pluton is hosted partly by the Napeu Kainiut quartz monzonite, a satellite 98 intrusion of the Mistastin batholith. Although the age of the Napeu Kainiut intrusion is 99 unknown, the Mistastin batholith has been dated at 1.42 Ga (Emslie and Stirling 1993). It is 100 one of the largest and oldest intrusions of the Nain Plutonic Suite, and covers a surface area 101 of about 5,000 km². The Mistastin batholith is composed dominantly of pyroxene- and 102 olivine-bearing Rapakivi type granites, which are cut by younger biotite-hornblende-bearing

granites. It is host to several smaller REE-rich intrusions, notably the Misery Lake syenite at 1.41 Ma (Petrella et al. 2014) and the Ytterby 2 granite and Ytterby 3 syenite-granite dated at 1.44 and 1.42 Ga, respectively (Kerr and Hamilton 2014). The Napeu Kainiut quartz monzonite is a biotite-bearing monzonitic to granitic rock. It is coarse-grained, equigranular, consists mainly of quartz, K-feldspar, plagioclase and biotite, and is of metaluminous composition.

The other host to the Strange Lake pluton is an Archean to Paleoproterozoic gneiss complex, in the Core Zone (southeastern Churchill Province), composed of quartzofeldspathic augen-gneisses, banded biotite-gneisses and minor garnet-bearing para-gneisses and mafic gneisses. This part of the Core Zone represents a poorly constrained collage of at least three Archean to Paleoproterozoic crustal lenses and domains bounded by shear zones, which represent former sutures (James and Dunning 2000). The crustal rocks near Strange Lake are dominantly (meta-) volcanics (2.3 to 2.5 Ga) of the Mistinibi-Raude domain, crosscut by younger gabbroic rocks, and represent former accreted volcanic arcs and products of later rifting events (Girard 1990; Corrigan et al. 2016). The Archean blocks are exotic and do not belong to any of the larger Archean terranes of the Rae, Nain or Superior Provinces, as earlier believed (Corrigan et al. 2009; James and Duning, 2000). The crustal lenses in which Strange Lake is located, formed at middle to lower crustal levels (≥ 10 km depth) and underwent amphibolite to granulite facies metamorphism (Van der Leeden et al. 1990). They are underlain by an Archean (~2.7 Ga) tonalitic to metasedimentary gneiss package belonging to the Mistinibi Complex (Girard 1990; Van der Leeden et al. 1990).

The eastern foreland of the Core Zone underwent eastward subduction beneath the Archean Nain craton at 1.91-1.87 Ga, resulting in the N-S-trending Torngat orogen (1.87-1.84 Ga), the calc-alkaline Burwell Domain and a broad belt of Tasiuyak paragneiss, a former sedimentary wedge (Scott 1998; Wardle et al. 2002). The Torngat event was followed by eastwards

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subduction of the eastern foreland of the Archean Superior Province under the Core Zone
(1.84-1.82 Ga), and subsequent continent-continent collision (1.82-1.77 Ga), which produced
the N-S trending New Québec orogen (1.87-1.79 Ga). This also led to the emplacement of
large syn- and post-collisional intrusive bodies (see Fig. 1), notably the N-S trending calcalkaline De Pas and Kuujjuaq batholiths (Hoffman 1988; Wardle et al. 2002).

A number of authors (e.g., Blaxland and Curtis 1977; Myers et al. 2008) have argued that the Nain Plutonic Suite and the Gardar Province of Southwest Greenland belong to a larger igneous suite that was divided in two parts by the rifting event that created the Labrador sea during the late Mesozoic to early Tertiary (Royden and Keen 1980). It is important to note, however, that the onset of the magmatic activity responsible for the Nain Plutonic Suite (ca 1.46 to 1.24 Ga) preceded Gardar magmatism (ca 1.35 to 1.12 Ga). The latter produced a series of alkaline gabbros, granites and nepheline syenites plus several small carbonatites (Bailey 2001; Upton et al. 2003). Evidence that the two alkaline provinces were linked is provided by anorthosite xenoliths in the Gardar intrusions, which are compositionally similar to the large anorthosite complexes in Labrador (Bridgwater 1967). It is also noteworthy that the 1.276 Ga mafic dike swarms (Nain Dikes) and the 1.273 Ga Harp Dikes of the Nain Province, are remarkably similar to the 1.273 Ga 'Brown Dikes' in the Gardar Province (Cadman et al. 1993; Carlson et al. 1993; Bartels et al. 2015) . Further evidence that the two provinces constitute a single larger province is provided by the basement to the intrusions of the Gardar Province, which is composed of the calc-alkaline Julianehåb batholith (1.80-1.77 Ga). This intrusion was emplaced during and following the Paleoproterozoic Ketilidian orogeny (1.85 to 1.80 Ga), which is represented by the Makkovik province (orogeny 1.895 to 1.870 Ga) in Labrador (Garde et al. 2002).

151 The Strange Lake pluton

The Strange Lake peralkaline granitic pluton is located on the border of Québec and Labrador and intruded along the boundary between the Mesoproterozoic Napeu Kainiut quartz monzonite to the West and a Paleoproterozoic gneiss complex of the Core Zone, to the East (Fig. 2). The pluton consists of a series of peralkaline granitic intrusions forming a cylindrical intrusive body with a diameter of about 6 km. A radiometric U-Pb determination on zircon for the oldest unit, a hypersolvus granite, yielded an age of 1240 ± 2 Ma (Miller et al. 1997). The hypersolvus granite, so-named because of the occurrence of a single perthitic alkali feldspar, forms the center of the pluton, and is divided into a less evolved southern and a more evolved northern unit-(Siegel et al. in prep.), the latter evident from higher bulk Si, Zr, Nb, REE and lower Al concentrations (Table 1). The hypersolvus granite was succeeded by the most evolved unit, a transsolvus granite, which is distinguished from it by the presence of separate crystals of albite and K-feldspar in addition to perthite. This granite occupies most of the pluton, is heavily altered except in the center of the pluton and is host to REE/HFSE mineralized pegmatites that have been the target of recent exploration (Gysi and Williams-Jones 2013; Gysi et al. 2016). A dark grey porphyritic granite is present between the transsolvus granite and large bodies of wall rocks, and is interpreted as the quenched margin of the former intrusion (Siegel et al. in prep.). This granite is also represented by dark grey, fine-grained ovoid enclaves that frequently occur in the transsolvus granite. A ring-fault and a fluorite-hematite breccia coincide with the margins of the pluton. The units referred to above and their evolution are described and discussed in more detail in Siegel et al. (in prep.).

172 METHODS

173 Samples

174 Twenty two samples were collected and prepared for Sm-Nd isotope analyses. These include175 seven bulk rock samples from the unaltered center of the Strange Lake pluton (four samples

of hypersolvus granite, two of transsolvus granite and one of quench zone granite) and five bulk rock samples from the Napeu Kainiut quartz monzonite host rock. Three of the host rock samples were collected within the borders of the pluton from large outcrops interpreted to represent roof pendants, and two samples were from outside the pluton (see Fig. 2). In addition, ten samples of sodic amphibole (arfvedsonite) were separated manually from the Strange Lake granites with the aid of a binocular microscope. Four of these separates are from samples that were analyzed for their bulk rock Sm-Nd isotopic composition. The other six separates were prepared from samples not subjected to this analysis, because arfvedsonite grains in the remaining samples that had been analyzed for their Nd-Sm isotopic composition contain abundant inclusions of fluorite and other minerals. This is a common feature of arfvedsonite in the hypersolvus granite. Quartz crystals from six granite samples (three of hypersolvus granite, one of quench zone granite, and two transsolvus granite samples) and two samples of quartz monzonite (one from inside, and the other from outside the Strange Lake pluton) were separated manually for oxygen isotope analysis; quartz is considered to best reflect magmatic δ^{18} O values in granitic intrusions (Gregory et al. 1989). Samples of the Strange Lake bulk rock were not considered for oxygen isotope analyses because of the high probability that late stage hydrothermal fluids affected the δ^{18} O values of the feldspar. Three samples of biotite gneiss, including a garnet-bearing variety, from the gneiss complex partly hosting the Strange Lake pluton were analyzed for their bulk rock oxygen isotope composition. One of the samples was collected about 1 km north of the pluton and the other two from wall rock outcropping immediately to the north of the pluton. The gneiss samples were prepared (crushed and ground) and analyzed at the Queen's University Facility for Isotope Research. The bulk rock samples used for the Nd isotope analyses were crushed and ground to fine powders using a jaw crusher and tungsten carbide mill at McGill University, whereas the arfvedsonite and quartz separates were crushed and ground manually in agate

201 mortars. The bulk-rock chemical data were provided by Quest Rare Minerals Ltd., and 202 represent the results of analyses by Actlabs using the fusion ICP-MS technique for the 203 determination of the REE concentrations.

204 Analytical methods

205 Radiogenic isotopes

All radiogenic isotope analyses were performed at GEOTOP (Université de Québec à Montréal, UQAM). About 40 - 80 mg of powder of each sample was spiked with a ¹⁵⁰Nd-¹⁴⁹Sm tracer solution in order to accurately determine Sm and Nd concentrations through isotope dilution. The rock powders were dissolved in acidic HF and HNO₃ solutions on a hot plate for six days. In order to decompose insoluble fluorides, HNO₃ acid was added and the solutions evaporated repeatedly. The elements, Sm and Nd, were separated by ion-exchange chromatographic techniques under clean laboratory conditions using doubly distilled acids. Iron was removed using ion-exchange polyprep columns with AG1X8 resin. The rare earth elements (REE) were concentrated using Eichrom TRU spec resin, and Sm and Nd were subsequently separated using columns prepared with two ml of Eichrom LN spec resin.

The samples were loaded onto Re-filaments and analyzed on a thermal ionization TRITON PLUS mass spectrometer (TIMS). A double filament assemblage was used, in which each sample was evaporated from the Re-filament on the left side of the sample wheel and ionized by a blank counterpart Re-filament on the right side. Both Sm and Nd were analyzed in static mode. The ¹⁴³Nd/¹⁴⁴Nd values were corrected internally for fractionation using a ¹⁴⁶Nd/¹⁴⁴Nd value of 0.7219, assuming exponential fractionation behavior. Repeated measurements of the Nd standard, JNdi, returned a mean value of 0.512097 \pm 7 (n = 5, 2 σ mean), which is comparable to the published value of 0.512115 ± 7 (Tanaka et al. 2000). Initial ¹⁴³Nd/¹⁴⁴Nd values for the Strange Lake samples were calculated using a U-Pb zircon age of 1.24 Ga

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(Miller et al. 1997), whereas the 1.42 Ga age of the Misastin batholith (Emslie and Stirling 1993) was assumed for the Napeu Kainiut quartz monzonite samples. The Sm-Nd concentrations were determined using the isotope dilution technique. The ENd values were calculated from the measured ¹⁴³Nd/¹⁴⁴Nd ratios relative to the chondritic uniform reservoir value (CHUR) using the present-day ratios of 143 Nd/ 144 Nd = 0.512638 (Goldstein et al. 1984) and 147 Sm/ 144 Nd = 0.1967 (Jacobsen and Wasserburg 1980). The average total chemical blank concentration measured on the TIMS was 150 pg for Nd and Sm, which is considered negligible.

233 Oxygen isotopes

The oxygen isotope analyses (n = 9) were carried out at the Laboratory for Stable Isotope Science (LSIS) at the University of Western Ontario (UWO) using a method similar to that described by Polat and Longstaffe (2014). Approximately 8-10 mg of powder from each sample was weighed into spring-loaded sample holders and dried overnight at ca. 150°C. The holders were then placed in nickel reaction vessels and heated in vacuo for a further three hours at 300°C to remove surface water. The samples were subsequently reacted with ClF₃ overnight at ca. 580°C to release silicate-bound oxygen (Borthwick and Harmon 1982; Clayton and Mayeda 1963). The oxygen was converted to CO₂ over red-hot graphite for isotopic measurement using a Prism II dual-inlet, stable-isotope-ratio mass-spectrometer. The oxygen isotope measurements were made using a Thermo Fisher Delta V Plus mass spectrometer in dual inlet mode. The gneiss samples (n = 3) were analyzed for their bulk rock δ^{18} O compositions at the Queen's Facility for Isotope Research (Queens University). Oxygen was extracted from 5mg samples at 550-600°C using the BrF5 procedure of (Clayton and Mayeda 1963) and analyzed in dual inlet mode with a Thermo-Finnigan DeltaPlus XP Isotope-Ratio Mass Spectrometer.

Oxygen isotope compositions are reported in the standard delta (δ) notation in units of permil (%), expressed relative to the Vienna Standard Mean Ocean Water (VSMOW) international standard. All samples were analyzed with a precision of 0.1‰. The δ^{18} O values for laboratory standards analyzed at the same time as the Strange Lake guartz samples were: guartz, $11.5 \pm$ 0.24 ‰ (n=5); basalt, 7.5 \pm 0.25 ‰ (n=4); and CO₂, 10.3 \pm 0.05 ‰ (n=5). The accepted values for these standards are 11.5 ‰, 7.5 ‰ and 10.3 ‰, respectively. The accuracy for the gneiss bulk rock values was 0.1 ‰, based upon primary or secondary standard analyses.

RESULTS

Rare earth element contents

Chondrite-normalized (Sun and McDonough 1989) REE-profiles for the Strange Lake bulk rock samples used in the Sm-Nd isotope study all display modest LREE enrichment, a strong negative Eu-anomaly and a flat HREE distribution (Fig. 3). The absolute REE concentrations are lowest in the least evolved southern hypersolvus granite, considerably higher in the northern hypersolvus granite and highest in the most evolved transsolvus granite (almost twice as high as in the southern hypersolvus granite; Table 2). The chondrite-normalized REE profiles of the Napeu Kainiut quartz monzonite samples display very similar LREE enrichment to the Strange Lake granites and absolute LREE concentrations similar to those of the southern hypersolvus granite. In contrast, the HREE are relatively depleted and the absolute HREE concentrations considerably lower than those of the Strange Lake granites. Interestingly, the quartz monzonite samples collected from within the Strange Lake pluton (roof pendants) have higher REE contents than the samples from outside the pluton.

Sm-Nd isotope geochemistry

Bulk rock concentrations of Sm and Nd for the Strange Lake granites range from 10 to 61 ppm and from 45 to 291 ppm, respectively (Table 3). They are lowest in the hypersolvus and quench zone granites (Sm: 10 - 40 ppm and Nd: 45 - 188 ppm) and highest in the transsolvus granite (Sm: 50 – 61 ppm and Nd: 291 - 310 ppm), reflecting the more evolved nature of the latter unit (Fig. 4). Samarium and neodymium concentrations in the arfvedsonite samples show no obvious trends with granite evolution, and range from 4 to 27 ppm and 19 to 121 ppm, respectively. This is probably because arfvedsonite was a late crystallizing mineral in the hypersolvus granite and crystallized as an early phase in the transsolvus granite. The Napeu Kainiut quartz monzonite samples have significantly lower Sm and Nd concentrations of 14 to 18 ppm, and 70 to 107 ppm, respectively, with the exception of sample 10120, which contains 28 ppm Sm and 120 ppm Nd, and was collected inside the Strange Lake pluton.

The ¹⁴⁷Sm/¹⁴⁴Nd ratios of the unaltered Strange Lake granites and arfvedsonite separates range from 0.0967 to 0.1654 (most values are clustered between 0.124 and 0.146), and there is no discernible difference between the values for the hypersolvus granite and the transsolvus granite (Fig. 5). In contrast, the quartz monzonite values are lower, clustering between 0.1029 and 0.1220, except for sample 10120, which has a ratio of 0.1394. Note that values > 0.13 are unusual in plutonic rocks (De Paolo 1988). The altered and mineralized Strange Lake samples analyzed by Kerr (2015) have much higher ¹⁴⁷Sm/¹⁴⁴Nd ratios than the unaltered samples analyzed in this study, and range between 0.1683 and 0.3903. The measured ¹⁴³Nd/¹⁴⁴Nd compositions of the Strange Lake granites and the arfvedsonite separates (this study) range between 0.511748 and 0.512304, and do not distinguish hypersolvus granite from transsolvus granite. These ratios for the Napeu Kainiut quartz monzonite are all considerably lower, between 0.511411 and 0.511527, except for the one sample (10120) with anomalously high Sm and Nd concentrations (Fig. 5, Table 3). This sample has a ¹⁴³Nd/¹⁴⁴Nd ratio similar to that of the Strange Lake granites, of 0.511882.

Arfvedsonite separates of some of the bulk rock samples have ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁴³Nd/¹⁴⁴Nd ratios that differ considerably from those of their host rocks (Table 4), indicating a fractionation of Sm from Nd between the melt and arfvedsonite. For example, the bulk rock sample, 10010, has a ¹⁴⁷Sm/¹⁴⁴Nd ratio of 0.1280 and a ¹⁴³Nd/¹⁴⁴Nd ratio of 0.511935, whereas the arfvedsonite separate of the same sample has ratios of 0.1659 and 0.512263, respectively.

The ε Nd (t) values for the Strange Lake samples were calculated using the crystallization age of the pluton of 1.24 Ga (Miller et al. 1997). These values vary considerably, from - 0.6 to -5.7 for the bulk rock samples, and from -0.3 to -5.3 for the arfvedsonite separates (Table 3). However, the majority of the values of both bulk rock samples and arfvedsonite separates lie between -1.4 and -4. Similar results were reported recently by Kerr (2015) for bulk rock samples from the Labrador side of the Strange Lake pluton. His values lie between -1.5 and -5.5, with the majority being between -2 and -3. Although the age of the Napeu Kainiut quartz monzonite is unknown, as noted above, the intrusion is widely considered to be a satellite of the nearby (and compositionally similar) Mistastin batholith. Thus, the age of the latter (1.42 Ga) was used to calculate the ε Nd (t) values for the quartz monzonite samples. They are between - 6.6 and - 8.1, except for sample 10120, which has a ENd value of -4.8 (Table 5). If the age of the Strange Lake granite (1.24 Ga) was used instead, the εNd (t) values would be more negative by about two units ($\epsilon Nd = -8.6$ to -9.9 and -6.8 for sample 10120).

Oxygen isotope ratios

The oxygen isotope ratios (δ^{18} O) for quartz from the Strange Lake samples (Table 4) are lowest for the least evolved southern hypersolvus granite (8.2 and 8.4 %), higher for the quench zone granite and the more evolved northern hypersolvus granite (8.4 and 8.7 ‰) and highest for the transsolvus granite (8.8 and 8.9 ‰). These values largely overlap with three

320 oxygen isotope values determined by Boily and Williams-Jones (1994) for quartz separates 321 from the northern hypersolvus granite (8.5 and 8.6 ‰) and the transsolvus granite (9.1 ‰). 322 The quartz monzonite quartz samples have slightly higher δ^{18} O values of 8.9 and 9.1 ‰. The 323 bulk rock biotite-gneiss samples have δ^{18} O values of 8.6 and 9.6 ‰, and the garnet-biotite-324 gneiss sample has a δ^{18} O value of 6.3 ‰.

DISCUSSION

326 The isotopic signature of the Strange Lake pluton

The fact that the hypersolvus and transsolvus granites cannot be distinguished on the basis of their ϵ Nd and δ^{18} O values, -1.4 to -4.1 and 8.2 to 9.1 ‰, respectively, suggests that the magmas responsible for these two granite types originated in the same reservoir. This means that the geochemical differences between the hypersolvus and transsolvus granites did not result from an external process like crustal assimilation, but instead from internal magmatic processes such as fractional crystallization. The bulk rock Sm and Nd concentrations are linearly distributed (see Fig. 4), consistent with an evolution of the magmas by fractional crystallization, i.e., they are lower in the hypersolvus granite and higher in the transsolvus granite. The highest concentrations of these elements, however, are in the altered rocks (see Fig. 6a), reflecting the important role of hydrothermal fluids in mobilizing the REE (Salvi and Williams-Jones 1996; Gysi et al. 2016).

Variations in the Sm/Nd ratios among the Strange Lake samples resulted either from fractionation induced by the crystallization of LREE-bearing minerals, hydrothermal activity or a combination of the two processes. The main reservoirs of Sm and Nd in the unaltered Strange Lake granites are monazite-(Ce), fluor-natropyrochlore and arfvedsonite (Siegel et al. in prep.; Siegel et al. in review). Experimentally determined partition coefficients between monazite-(Ce) and melt are consistently higher for Nd than for Sm over a wide range of

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344 pressure-temperature conditions and melt-H₂O contents (Stepanov et al. 2012). The same is 345 true for Nd and Sm partition coefficients between pyrochlore group minerals and silicate 346 melts (Zhao et al. 2002). Arfvedsonite, a major phase in the granite (~15 vol. %), also has a 347 slight preference for Nd over Sm (calculated D_{arf-melt} Nd ~0.009 vs. D_{arf-melt} Sm ~0.008) 348 (Siegel et al. in review). Thus, all primary (magmatic) REE-bearing minerals at Strange Lake 349 preferentially incorporated Nd over Sm.

Evidence that the magmatic Sm/Nd signature was affected by hydrothermal activity is provided by quartz monzonite sample #10120, which was collected from a Napeu Kainiut roof pendant located in close proximity to a mineralized Strange Lake pegmatite dike. This sample underwent strong hydrothermal alteration, resulting in anomalously high total Rare Earth oxide (0.4 wt. %), F (1.4 wt. %) and CaO (2.7 wt. %) concentrations, whereas the other quartz monzonite samples, which were only weakly altered (samples from within the Strange Lake pluton) or not altered at all (samples from outside the pluton) have very low total Rare Earth oxide (0.06 - 0.09 wt. %), F (0.07 - 0.17 wt. %) and CaO (0.75 - 1.68 wt. %) contents. Therefore, we conclude that the much higher Sm and Nd concentrations of sample #10120 compared to those of the other quartz monzonite samples, as well as its high ¹⁴⁷Sm/¹⁴⁴Nd ratio (0.1394), were likely the result of interaction with REE-rich fluids that were exsolved from the nearby Strange Lake pegmatite. These fluids must have had higher Sm/Nd ratios than the unaltered rock. Kerr (2015) also noted that highly mineralized samples (e.g., aplites, pegmatites) from the Labrador side of the Main Zone deposit had ¹⁴⁷Sm/¹⁴⁴Nd ratios as high as 0.1683 to 0.3903 (see Fig. 6b). These mineralized samples show evidence of intense hydrothermal alteration, and the REE are hosted dominantly in secondary minerals, such as gerenite-(Y), kainosite-(Y) and gadolinite-(Y) (Kerr and Rafuse 2012). The high ¹⁴⁷Sm/¹⁴⁴Nd ratios indicate that the Sm-Nd system in these altered samples is highly disturbed. Indeed, they result in depleted mantle model ages (T_{DM}) that are younger than the intrusion or even

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lie in the future (Kerr, 2015). Mass-balance calculations and geochemical modeling suggest that low-temperature hydrothermal fluids preferentially mobilized the LREE over the HREE (Salvi and Williams-Jones 1996; Gysi and Williams-Jones 2013; Gysi et al. 2016). A fluid containing both Sm and Nd would preferentially enrich an unaltered rock in the slightly 'heavier' and less mobile Sm, whereas Nd is lighter and would be transported further (Migdisov et al. 2016). This explains the unusually high ¹⁴⁷Sm/¹⁴⁴Nd ratios of the altered granites and pegmatites.

376 Model ages

Model ages are a function of the ¹⁴⁷Sm/¹⁴⁴Nd ratio and the model used for the mantle (depleted vs. enriched). As the ¹⁴⁷Sm/¹⁴⁴Nd ratios of the Strange Lake granites are highly variable, and as choice of the mantle model is made difficult by the fact that the composition of the mantle beneath Strange Lake is unknown, ages calculated for this mantle should be treated with caution. Applying the depleted model (T_{DM}) would require that there had been extensive melting of the mantle prior to the alkaline magmatism. The enriched or T_{CHUR} (chondritic evolution) model (De Paolo and Wasserburg 1976) would require the assumption that the mantle beneath Strange Lake had not experienced appreciable depletion (e.g., by melting to produce crust) prior to the alkaline magmatism or that it had been enriched metasomatically.

The calculated depleted model ages (T_{DM}) for the full Strange Lake dataset are between 1.77 and 3.18 Ga. However, in order to exclude samples that may have been affected by nonmagmatic Sm-Nd fractionation, we restricted the data to ¹⁴⁷Sm/¹⁴⁴Nd ratios < 0.13 (De Paolo 1988). This restriction reduced the T_{DM} range to between 1.77 and 2.20 Ga (**Fig. 7**). For reference, the corresponding range of enriched mantle model ages (T_{CHUR}) (De Paolo 1988) is between 1.27 and 1.74 Ga. The variability in both the depleted and enriched mantle model

ages indicates that the Strange Lake samples were highly affected by fractionation. In addition the samples were also affected by crustal contamination, as shown by the relatively low initial 143 Nd/ 144 Nd and ε Nd values.

The Napeu Kainiut quartz monzonite samples have depleted mantle model ages (T_{DM}) from 2.38 to 2.69 Ga, whereas their enriched mantle model ages (T_{CHUR}) are between 1.99 and 2.26 Ga, both of which are significantly older than the model ages for Strange Lake (Fig. 7). For the reasons given above, the model ages and their geological significance should be treated with caution. Nonetheless, the large gap in model ages (enriched and depleted) predicted for the mantle by the Napeu Kainiut quartz monzonite and Strange Lake granites is significant and suggests that the two rock suites followed separate evolutionary paths. This is further elaborated upon below.

Relationship of the Strange Lake granites to the Napeu Kainiut quartz monzonite

The quartz monzonite samples from within and outside the Strange Lake pluton have lower ϵ Nd (-4.8 to -8.1, at t=1.42 Ga) and slightly higher δ^{18} O (8.9, 9.1 ‰) values than most of the Strange Lake samples. These differences can be explained by a higher degree of crustal contamination for the Napeu Kainiut pluton or its formation from crustal melts. The similarity in the mineralogy of the Napeu Kainiut pluton to that of the 1.42 Ga Mistastin batholith (Miller et al. 1997), and obvious differences in their isotopic signatures from those of the Strange Lake granites, as noted above, suggests that the magmas forming the Strange Lake pluton followed a different evolutionary path from those producing the former intrusions. In addition, the model ages reveal that initial melt extraction from the mantle may have been much later for the Strange Lake granites than for the rocks of the Napeu Kainiut pluton (Fig. 7). It is important to note, however, that this does not exclude the possibility that both intrusions were the products of the same large-scale tectonic processes or had the same

primary magma source albeit with a greater mantle contribution in the case of the Strange Lake granites. Indeed, a common source is supported by the similarity of the shapes of the chondrite-normalized REE profiles of the Napeu Kainiut samples from outside the Strange Lake pluton to those of the Strange Lake granites. However, the Eu anomalies of the latter are stronger and their HREE trends flatter (see Fig. 3). Theoretically, it is possible that the extreme REE, Zr, Nb and alkali enrichment of the Strange Lake pluton could have originated from the melting of crustal material. However, on the basis of the data presented, we suggest that it is much more likely that this enrichment was due to the involvement of a mantle melt or, as suggested by Martin (2006), metasomatism of the crust by mantle fluids prior to melting. A large number of isotopic studies of alkaline suites (e.g., Kerr and Fryer 1993; Stevenson et al. 1997; Kovalenko et al. 2009; Hegner et al. 2010) have concluded that the trace element enrichment of these suites was the product of combined crustal contamination and fractional crystallization of mantle-derived melts. On the basis of our data, we propose a similar origin for the silica-oversaturated Strange Lake pluton, which is examined in the following sections. However, the formation of the pluton from crustal melts metasomatically altered by mantle fluids (Martin 2006) cannot be ruled out, as granitoid rocks produced in this manner would have isotopic signatures that would be indistinguishable from those produced by crustally contaminated mantle melts.

435 Nature of the mantle underlying the Core Zone

In order to evaluate the impact of crustal processes on the evolution of the Strange Lake pluton from a magma of mantle origin, it was first necessary to gain insights into the nature of the underlying mantle, which elsewhere in the Core Zone has been described as being either depleted or enriched (Ashwal et. al 1986; Amelin et al. 2000; Bedard 2001). Important insights into this nature can be gained from the behavior of the REE, which depends on the presence (or absence) of minerals that can sequester them and whether these minerals

preferentially accept the LREE or the HREE. Most of the intrusions of the Nain Plutonic Suite (e.g., Emslie et al. 1994; Petrella and Williams-Jones 2014), including the Strange Lake granites, have chondrite-normalized REE profiles, which show some LREE enrichment, a pronounced negative Eu-anomaly and a flat HREE pattern (see Fig. 3). Thus, the mafic rocks (olivine gabbro, troctolites, gabbroic anorthosites) have chondrite-normalized La/Lu_N ratios between 0.8 and 1.0 (Emslie et al. 1994), and the least evolved Strange Lake granites have La/Lu_N ratios between 3 and 5. These low Lu/La ratios rule out a garnet-bearing mantle, because low degrees of partial melting of such a mantle would produce melts enriched in the LREE and depleted in the HREE due to retention of the latter by garnet (De Paolo 1988). For comparison, low degrees of melting (1-3 %) of a garnet lherzolite would produce La/Lu_N ratios between ~200 and 400 in the resulting magma (calculated assuming a mantle composed of 50 % olivine, 30 % orthopyroxene, 10 % clinopyroxene and 10 % garnet and using mineral-melt partition coefficients from De Paolo (1988)). The REE profiles for the Nain Plutonic rocks, including the Strange Lake granites, are thus consistent with fractionation of a parental melt from a garnet-poor lherzolite or garnet-free lherzolite at depths less than those for which garnet is stable; < 85 km (Robinson and Wood 1998). Moreover, such shallow melting could be promoted by the addition of water from subducted oceanic crust (Green 1973). The isotopic compositions of olivine-bearing mafic intrusions and large mafic dike swarms exposed in the region should, in principle, be closest to those of the underlying mantle (higher degrees of partial melting). The 1.33 Ga mafic Voisey's Bay intrusions, which lie about 150 km to the east of Strange Lake, are some of the most mafic intrusions of the Nain Plutonic Suite, and thus closest in composition to mantle (Amelin et al. 1999). The least contaminated samples are olivine-gabbros and troctolites that have ENd values from +0.9 to -2.1 (Table 5) and have been interpreted to represent either melting of an enriched subcontinental mantle or a depleted mantle contaminated by minor proportions of crustal

material (Amelin et al. 2000). The olivine-bearing basaltic 'Nain Dikes' of the Nain Plutonic Suite, with ε Nd values of -0.2 and -1.2 and δ^{18} O values of 6.6 and 6.8 ‰, also have been interpreted to represent an enriched mantle source (Carlson et al. 1993). Carson et al. (1993), however, did not entertain the possibility of a depleted mantle contaminated by crust.

As noted earlier, the Nain Plutonic Suite is interpreted to belong to a larger igneous province that includes SW-Greenland. Thus, studies of the Gardar Province could provide insights into the nature of the mantle beneath the Nain Plutonic Suite. Stevenson et al. (1997) attributed the ɛNd variations in the Gardar igneous rocks to crustal contamination of a depleted mantle. This interpretation was based on ε Nd values of +2.1 to +2.4 for the Eriksfjord basalts (Paslick et al. 1993) and a EHf value of +3.2 for an eudialyte sample from Ilímaussaq (Patchett et al. 1981). Negative ε Sr values and ε Nd values between +1 and +5 for the Igaliko carbonatite and associated lamprophyre dikes also suggest a depleted mantle (Pearce and Leng 1996; Halama et al. 2003). However, phonolites and oversaturated rocks of the same suite have signatures indicative of crustal contamination, such as strongly positive ESr values (26 and 115), only slightly positive ε Nd values (1.2 and 1.3) and a wide range in δ^{18} O values (6.1 to 15.9 ‰). In contrast, mafic dikes in the vicinity of the main intrusions of the Gardar Province (e.g., the 1.276 Ga Brown Dikes) with ε Nd values between -4.4 and -0.5 and a relatively flat HREE profile have been interpreted to originate from an enriched mantle that underwent partial melting at depths less than those of the garnet stability field (< 85 km) (Bartels et al. 2015). Other evidence of an enriched lithospheric mantle is provided by the occurrence of peridotitic mantle xenoliths (Igdlutalik) with high Zr and Nb contents and low Zr/Nb values (< 4) (Upton 1991). Significantly, ultramafic lamprophyres and carbonatites in the younger southern Gardar rift have similarly low Zr/Nb (< 4) ratios (Upton et al. 2003; Upton 2013). In order to reconcile the apparently contradictory conclusions over whether the mantle underlying the Gardar Province is depleted or enriched, a number of researchers have argued

for a mantle that initially was broadly similar (depleted) but became heterogeneous due to locally variable degrees of mantle metasomatism (Goodenough et al. 2002; Upton et al. 2003; Bartels et al. 2015). This interpretation of metasomatism of the lithospheric mantle beneath the Gardar Province is based on trace element concentrations that differ from common intraplate OIB (ocean island basalt) distributions, and are ascribed to the infiltration of subduction-related alkali and volatile rich fluids associated with the 1.80 Ga Ketilidian orogeny (Goodenough et al. 2002; Upton 2013).

Evidence of the Ketilidian orogeny is absent in northern Québec-Labrador, with the exception of the Makkovik Province (Kerr and Fryer 1994). However, the alkaline magmatism of the Nain Province was preceded by the eastward subduction of the foreland of the Superior craton under the Core Zone in the Paleoproterozoic at 1.84 to 1.82 Ga (Van Kranendonk et al. 1993; Wardle et al. 2002). In both northern Québec-Labrador and southern Greenland, there was enrichment of the subcontinental lithospheric mantle through subduction of an ocean floor that contributed to the extreme enrichment in F, alkalis and REE of the Gardar and Strange Lake intrusions (Goodenough et al. 2002; Upton 2013). We conclude, on the basis of the evidence presented for both the Gardar and Nain Provinces that the mantle underlying the Strange Lake pluton was initially depleted and subsequently enriched by metasomatic fluids produced by subduction-related melts.

The composition of the crust beneath Strange Lake

As discussed earlier, the isotopic data provide compelling evidence that the Strange Lake magmas experienced crustal contamination. Here we evaluate possible sources of this contamination in the basement hosting the pluton, i.e., in the reworked Archean and Paleoproterozoic crustal lenses that constitute the Core Zone.

Seismic surveys and gravity data indicate that the crust of the Core Zone has a thickness of 35 to 40 km, of which the middle to lower part is composed mostly of refractory Archean lithosphere (Wardle et al. 2002). The upper crust of the Core Zone is composed of largely undifferentiated Archean to Paleoproterozoic gneisses of amphibolite to granulite facies (Wardle et al. 2002), which appear as separate blocks, commonly of metavolcanic arc material that are separated by shear zones (Girard 1990; James et al. 1996). The gneissic rocks in the proximity of the Strange Lake pluton consist dominantly of Paleoproterozoic gneisses, such as banded biotite paragneiss and quartzofeldspathic (augen-) orthogneiss and subordinate proportions of garnet-biotite paragneiss and mafic gneisses.

Only the quartzofeldspathic orthogneisses in the vicinity of Strange Lake have been analyzed for Sm-Nd isotopes and only the biotite-gneisses for oxygen isotopes. The quartzofeldspathic (granitoid) gneisses have an average ENd value of -12.7 at 1.24 Ga (Kerr 2015), and the biotite- (and one garnet-biotite-) gneisses have δ^{18} O values between 6.3 and 9.6 % (this study). Except for a single sample of biotite gneiss, these δ^{18} O values of the gneisses are lower than those for the Strange Lake granites; the exception has a δ^{18} O value of 9.6 ‰ (Table 5). This therefore likely rules them out as contaminants of the Strange Lake magma. Other Paleoproterozoic gneisses further from Strange Lake, for which oxygen isotopic data are available, are the Tasiuyak paragneisses located about 100 km to the east, which have δ^{18} O values between 8.3 and 16.1 %. This unit would have been a plausible contaminant, if it had been present in the vicinity of Strange Lake.

The Archean component of the western Core Zone at middle to lower crustal levels (> 6-10 km depth) is represented by the Mistinibi complex, which consists of intercalated ~2.7 Ga old tonalitic gneiss and paragneiss (Girard 1990; Van der Leeden et al. 1990; Wardle et al. 2002).
The Mistinibi complex, which underlies the Core Zone in the Strange Lake region, is a plausible contaminant because it is predicted to have strongly negative ɛNd values due to its

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540 Archean age, and positive δ^{18} O values due to the large sedimentary component. This proposal 541 is supported by the observation that the De Pas batholith, located about 50 km to the west of 542 Strange Lake, has similar ϵ Nd values to the Strange Lake granites (-3 to -7), and is 543 interpreted to have experienced significant contamination by the same Archean crustal 544 material (Kerr et al. 1994; Wardle et al. 2002).

545 Modeling the evolution of δ^{18} O and ϵ Nd

The extent of crustal assimilation by the Strange Lake magmas is evaluated here using a combined fractional crystallization and assimilation (AFC) model applied to the δ^{18} O and ENd data. Oxygen isotope compositions provide a reliable means for evaluating magma sources because they are relatively unaffected by the degree of partial melting or extent of fractional crystallization (Taylor 1978). Mantle rocks, whether depleted or enriched, have a δ^{18} O value of 5.7 ± 0.2 ‰ (Ito et al. 1987). Thus, magmatic rocks originating from the mantle with values of δ^{18} O >7 are interpreted to reflect assimilation of crust, with the higher values corresponding to higher proportions of sediment-derived material (see Fig. 8) (Taylor 1978). The oxygen isotopic fractionation between magma and silicate minerals is small at magmatic temperatures (1 > α > -1). A value of 0.8 for was calculated for the fractionation between quartz and rhyolitic melt ($\Delta^{18}O_{quartz-melt}$) at 750°C using the empirically derived equation of Zhao and Zheng (2003).

We employed the AFC model of De Paolo (1981) to evaluate the combined effects of fractional crystallization and the assimilation of wall rock on the isotopic evolution of the Strange Lake granites. In this model, the ratio between the mass assimilation rate (M_a) and fractional crystallization rate (M_c) is defined as 'r', where M_a in common igneous systems is a fraction of M_c (De Paolo 1981). We assumed, in the case of the oxygen isotope data, that the δ^{18} O value of the least contaminated Voisey's Bay mafic intrusion (6.3 ‰) was the

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starting value for the magma and assigned values for the contaminant that could explain the \hat{o}^{18} O values of the Strange Lake granites. As is evident from Figure 9a, only contaminants with δ^{18} O values ≥ 10 ‰ can explain the δ^{18} O values of the Strange Lake granites. Moreover, a contaminant of 10 %, would require that the 'r' value be > 0.6, which is unrealistically high (De Paolo 1981). In order to be able to use a more reasonable value of 'r', for example 0.3 or 0.4, it would be necessary to call on a contaminant with a δ^{18} O value of ~ 11 ‰ (Fig. 9a). Such a contaminant would produce the desired range of δ^{18} O values (8.2 – 9.1 ‰) after between 60% and 85% crystallization. Clearly, the ô¹⁸O values of the biotite gneisses in the immediate vicinity of the Strange Lake pluton are too low for them to have been the contaminant; the maximum value obtained for these gneisses was 9.6 ‰ (Table 4). On the other hand, Churchill Province Paleoproterozoic paragneisses of the Tasiuyak formation near Voisey's Bay, which have δ^{18} O values between 8.3 and 16.1 ‰ and an average δ^{18} O value of 10.9 ‰, would have been ideal contaminants (Table 5). As noted above, however, we consider it more likely that the contaminant was an Archean paragneiss of the Mistinibi Complex, which is thought to underlie the Strange Lake pluton. Unfortunately, the oxygen isotope data required to evaluate this possibility are not available. For comparison, simple fractional crystallization without contamination leads to a small decrease in δ^{18} O (**Fig. 9b**).

In the case of the Nd isotope data, the very small coefficients ($D_{Nd} \ll 1$) for the partitioning of Nd between early crystallizing minerals (olivine, pyroxenes and plagioclase) and the melt, allow the distribution coefficient (D) to be excluded from the AFC equations of De Paolo (1981). The Nd melt concentration (starting at 40 ppm, the average composition of olivinebearing basaltic dykes in the Nain Province, Carlson et al. 1993) thus increases exponentially with crystal fractionation, even though a significant proportion of crustal material (5, 10, 15 and 25 %) with a slightly lower Nd concentration (30 ppm, see below) is assimilated (Fig. 10). As starting compositions for the combined ε Nd - δ^{18} O AFC model, we used a ε Nd value

of +0.9 and a δ^{18} O value of 6.3 ‰, which represent the highest ϵ Nd composition, and lowest δ^{18} O composition, respectively, of the least contaminated mafic rock series at Voisey's Bay (Amelin et al. 2000) and olivine bearing basaltic dikes in the Nain Province (Carlson et al. 1993) (Tables 4; 5). Crustal material with a δ^{18} O value of 11 ‰ was used as the contaminant (see previous paragraph). Data published on Core Zone Paleoproterozoic to Archean gneisses provided the ENd isotopic composition (avg. -15) (Emslie et al. 1994) and Nd-concentration (avg. 30 ppm) (Kerr and Fryer 1993; Amelin et al. 2000; Kerr 2015) of the contaminant. The 'r' value was set at 0.35 (this value vields the observed Strange Lake δ^{18} O values and a realistic degree of fractional crystallization, see above) and the proportions of assimilated crustal material were fixed at 5, 10, 15 and 25 %. The parameters and equations used for the mixing calculations are reported in the appendix and in Table A1.

Four mixing scenarios were evaluated (Fig. 11). Mixing of a melt of basaltic dike composition with 5, 10 or 15 % of contaminant material produced the ϵ Nd and δ^{18} O values determined for the Strange Lake granites (pink box in Fig. 11). In order to model the isotopic values for the Napeu Kainiut quartz monzonite, a higher proportion of assimilated crustal material (~25 %) was required (orange line in Fig. 11). Note that the evolution of the ε Nd values from slightly positive values to as low as -5 depends on the proportion of contaminant (up to 15 %), and to a much lesser extent on fractional crystallization, which has a negligible effect on the Nd-isotopic composition. In contrast, the δ^{18} O values increase independently of the contaminant proportions, and instead reflect the interplay between the rate of addition of contaminant and the extent of crystallization of the magma.

In summary, the Nd and oxygen isotopic compositions of the Strange Lake granites are satisfactorily explained by fractional crystallization of a reasonably enriched mantle melt that assimilated between 5 and 15 % crust represented by paragneiss with a highly positive δ^{18} O value (~ 11) and strongly negative ϵ Nd value. As neither the Paleoproterozoic gneisses in the

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vicinity of Strange Lake or gneisses elsewhere in the Core Zone (except for the Tasiuyak paragneiss) could be the source of this contamination, we consider it much more likely that rocks of the Archean Mistinibi tonalite-paragneiss complex underlying the central and western Core Zone were the source. This view is supported by the observation that the De Pas batholith is interpreted to have been contaminated by the latter rocks and has ENd values (-3 to -5) similar to those of the Strange Lake granites (Kerr et al. 1994; Wardle et al. 2002). Assimilation of additional upper crustal material is likely (e.g., biotite-paragneiss and quartzofeldspathic orthogneiss). However, because the isotopic signatures are similar to or lower than those of the Strange Lake pluton, such assimilation would not be evident in the isotopic record of the final product.

Plate tectonic controls on the formation of A-type granites

An environment conducive to the generation of mineralized A-type granitoid rocks, such as the Strange Lake pluton as well as other silica-saturated and undersaturated REE-rich rocks, develops as a result of the following tectonic processes: 1) subduction of basaltic (alkali- and volatile-rich) oceanic crust; 2) fertilization of subcontinental mantle with alkalis and volatiles (F, Cl, H₂O, CO₂); 3) continent-continent collision; 4) crustal thickening (orogeny) with the emplacement of syn- and post-collisional calc-alkaline plutons, and perhaps the delamination of a thickened lithosphere; and 5) crustal extension induced either by post-collisional de-stressing or other rifting processes that favor alkaline magmatism (failed rifts) (Black et al. 1985; Martin 2006; McLelland et al. 2010). The geological history of Laurentia during the Paleo- and Mesoproterozoic records this sequence of tectonic processes.

In the eastern foreland of the Superior Province, the above sequence began with Paleoproterozoic rifting at 2.2 to 2.1 Ga and possibly again at 1.88 Ga, leading to the generation of juvenile oceanic crust (Wardle et al. 2002) that was subducted eastwards under

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the Core Zone between 1.84 and 1.82 Ga and metasomatized the subcontinental mantle (Fig.
12a and b). The collision between the Superior province and the Core Zone resulted in the
New Québec orogen and produced large syn-and post-collisional 1.84–1.81 Ga calk-alkaline
intrusions (De Pas and Kuujjuaq batholiths) (Fig. 12c). This thickening of the crust exerted
significant pressure on the underlying lithospheric mantle, which resulted in a low degree of
partial melting.

The collisional events at ~ 1.82 Ga were followed by a long period of tectonic quiescence within the Core Zone until the beginning of the Nain magmatism at ~1.46 Ga. Partial melting began about 360 Ma after the collision, and was most likely induced by decompression of the mantle due to repeated rifting (Fig. 12d) in the waning stages of the Elsonian orogenic event (Gower and Krogh 2002). Although the Core Zone remained relatively quiescent, tectonic activity continued along the southern margin of the Superior and North Atlantic (e.g., Nain Province) cratons with the formation of a marginal basin and the accretion of magmatic arcs during pre-Labradorian (> 1.71 Ga) and Labradorian events (1.71-1.62 Ga) (Gower and Krogh 2002). This was followed by a change to north-trending flat subduction beneath the Core Zone during the Pinwarian (1.52-1.46 Ga) and Elsonian (1.46-1.23 Ga) orogenies that terminated with the Grenville collision at 1.08-0.98 Ga (Gower and Krogh 2002). The Elsonian event gave rise to at least three stages of large-scale magmatism related to postcollisional crustal extension. These produced the 1.46-1.41 Ga Mistastin, Harp Lake and Michikamau intrusions, the 1.35–1.29 Ga Nain plutons and the 1.29–1.24 Ga Harp and Nain dikes, as well as highly evolved Flowers River and Strange Lake peralkaline rhyolitic / granitic suites (Gower and Krogh 2002).

660 There is compelling tectonic and geochronological evidence that the plutonic activity of the 661 southern Nain Province was influenced by a mantle enriched through Paleo- and 662 Mesoproterozoic pre-Grenvillian northward subduction, such as the Makkovikian event at ca.

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1.80 Ga (Kerr and Fryer 1994) and the Pinwarian event at ca. 1.50 Ga (Gower and Krogh 2002). An argument could even be made for a multi-stage-fertilization of the mantle underlying the Core Zone, starting with the eastward subduction at ~1.82 Ga associated with the Superior Craton followed by the two above mentioned pre-Grenvillian subduction events. Although much of this activity occurred some 400 km to the south of Strange Lake, it is noteworthy, for comparison, that Andean volcanism occurs up to 500 km from the trench along the South American coast (Mamani et al. 2010).

670 Correlations between Strang Lake and other REE-mineralized intrusions

A number of smaller, mostly silica-saturated and REE-mineralized intrusions are either hosted by the 1.42 Ga old Mistastin batholith or are associated with the older Nain magmatism (1.45 to 1.41 Ga). These include the Ytterby 1, 2 and 3 intrusions (Kerr and Hamilton (2014) and the REE-rich Misery Lake ferro-syenite (Petrella et al. 2014).

The relative proximity of these REE mineralized bodies to each other suggests that they may be products of the same fertile source. The negative ɛNd values of the Strange Lake granites and those of a 1.42 Ga granite ($\epsilon Nd = -9.1$) and a 1.44 Ga quartz syenite ($\epsilon Nd = -8.8$) from REE-enriched intrusive bodies within the Mistastin batholith (Kerr and Hamilton 2014) indicate that all of these mineralized intrusions underwent significant crustal assimilation. However, the fact that most of the REE-rich bodies to the south are silica-saturated syenites, whereas Strange Lake is silica-oversaturated, could indicate that there was greater crustal assimilation towards the North (Kay and Gast 1973; Sobolev et al. 2007). The older ages of the Ytterby and Misery Lake intrusions (1.41 to 1.44 Ga) (David et al. 2012; Kerr and Hamilton 2014) compared to Strange Lake (1.24 Ga) may reflect thickening of the crust with time, allowing greater contamination and silica-saturation of the Strange Lake parental magma. Further work is required to constrain these possible relationships.
Similarities and differences between the Nain and Gardar igneous provinces

We have argued that the source for the alkaline magmatism in Labrador (Nain Plutonic Suite) was likely a heterogeneous, and partially enriched subcontinental mantle, and that fertilization of this mantle occurred during the subduction of the eastern Superior Province foreland beneath the Core Zone at 1.84 - 1.82 Ga. A similar conclusion can be drawn for the Gardar Province, where there is also evidence for a highly heterogeneous mantle characterized by locations that are enriched and locations that are depleted (Stevenson et al. 1997; Halama et al. 2003; Sobolev et al. 2007). Basaltic magmas emplaced early in the evolution of the Gardar Province have geochemical signatures of subduction-related melts, i.e., high LILE, LREE and low Nb concentrations (Goodenough et al. 2002; Köhler et al. 2009), which have been attributed to the 1.85-1.80 Ga Ketilidian subduction (Garde et al. 2002; Upton 2013).

On both sides of the present Labrador Sea, a tectonic quiescence of several hundred million years separated the subduction events from the large-scale alkaline magmatic periods. In the Gardar Province, more than 500 Ma elapsed between the Ketilidian subduction at ~1.8 Ga and the start of alkaline magmatism at ~1.3 Ga, whereas on the Labrador side, the interval was ≤ 400 Ma between the end of the Superior subduction at ~1.82 Ga and the start of Nain plutonism at 1.46 Ga. Both sides of the Labrador Sea experienced several stages of rifting along different axes, resulting in crustal thinning and low degrees of partial melting that produced small melt fractions with high volatile and incompatible element concentrations (e.g., Upton et al. 2003; Myers et al. 2008). The repetition of this rifting is particularly evident in the occurrence of several generations of mafic dikes in the Nain Province, some of which extend into the Gardar Province (Cadman et al. 1993; Bartels et al. 2015). The main stage of emplacement of the Nain Plutonic Suite (1.35-1.29 Ga) was controlled by a ~N-S trending zone of crustal weakness, parallel to the suture between the Archean Nain Province

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and the Core Zone (Myers et al. 2008). In contrast, the main intrusions of the Gardar Province, the Ilímaussaq and Tuutoq intrusions, are aligned along an old SW-NE trending lineament of lithospheric weakness (Upton et al. 2003). In addition, the large Gardar-Voisey's bay lithospheric-scale fault zone (**Fig. 13**) aligns with and may be related to the major phases of alkaline magmatism (e.g., the Strange Lake pluton) that were localized along an extension of this fault zone (Myers et al. 2008).

There are several important differences between the Nain and Gardar igneous provinces. For example, the Nain Province magmatism is overall slightly older (1.46 to 1.24 Ga) and less alkaline than the Gardar province, which was emplaced between 1.35 to 1.12 Ga and produced highly agpaitic rocks. The main phase of Nain magmatism (1.35 to 1.29 Ga), however, coincides temporally with the earlier phase of Gardar magmatism (1.35 to 1.30 Ga) (Upton et al. 2003; Myers et al. 2008). There are compositional differences between the two provinces with nepheline syenites predominating in the Gardar Province (Upton and Blundell 1978) and anorthosites, norites, ferrodiorites, monzonites and granites being prevalent in the Nain Province (Gower and Krogh 2002). The differences in compositions (e.g., degree of alkalinity and silica saturation) and timing of magmatism in the two provinces may be due to several factors. Any enrichment of the mantle underlying the two provinces would have resulted from separate subduction events, namely the Ketilidian (Gardar Province) and the Superior (Nain Plutonic Suite) subduction. The nature and extent of fertilization of the subcontinental mantle under each province would therefore likely have differed because of differences in the compositions of the subducted slabs. Also, the Ketilidian crust, e.g., the Julianehåb batholith (Gardar Province), and the crust of the Core Zone (Nain Plutonic Suite), which are both the hosts and the contaminants of the intruding melts, differ in their proportions of intrusive, volcanic and sedimentary rocks. In addition, crustal extension was not homogenous throughout Eastern Laurentia, as implied by extensive dike swarms of

r37 several generations and axial orientations. The similarities between the settings of the two
r38 igneous provinces, however, exceed the differences and, in both cases, the tectonic
r39 environment was favorable for the generation and emplacement of alkaline rocks highly
r40 enriched in the REE and other HFSE.

741 CONCLUSIONS

The Nd- and O-isotope ratios for the unaltered Strange Lake granites presented in this study are consistent and differ from those of the surrounding Napeu Kainiut quartz monzonites, indicating different evolutionary paths for the two intrusions. The isotopic signatures of the Strange Lake samples can be explained by a combination of crustal assimilation and in-situ Sm-Nd fractionation, involving the crystallization of primary LREE-minerals and arfvedsonite and later hydrothermal remobilization of the REE.

The numerous reports of rocks with enriched isotope signatures among otherwise uncontaminated mafic rocks of the Nain Plutonic Suite (Emslie et al. 1994; Amelin et al. 2000; Peck et al. 2010; Kerr and Hamilton 2014), suggest that the Strange Lake magma was derived from a mantle that locally was metasomatically enriched in incompatible elements. Fractional crystallization of this magma and accompanying assimilation of 5-15 % of Archean paragneiss yielded the ε Nd and δ^{18} O values preserved by the Strange Lake granites. Generation of REE-mineralized A-type granites, as well as other silica-saturated members of the Nain Plutonic Suite, was likely due to a combination of tectonic events, including the 1.84-1.82 Ga subduction of oceanic crust of the Superior province foreland, which metasomatized the subcontinental mantle. This subduction was followed by continentcontinent collision and the New Québec orogeny, which was accompanied by calc-alkaline magmatism. Rifting of the Nain Province during the waning stages of the Elsonian event

between 1.46 and 1.24 Ga produced alkaline melts which were generated via low degrees ofpartial melting.

The alkaline intrusions of the Gardar province in SW-Greenland are similar to the Nain Plutonic Suite in nature and timing, but metasomatism of the Gardar mantle was related to the 1.85-1.80 Ga Ketilidian subduction and the Core Zone mantle to the 1.84 - 1.82 Ga Superior subduction. In addition, the underlying crust and main contaminant of the Gardar suites was the Paleoproterozoic calc-alkaline Julianehåb batholith, whereas for the Nain Plutonic Suite it was a highly heterogeneous mixture of Archean and Paleoproterozoic crust. A link between the two provinces is indicated by the extensive supra-regional dike swarms (Nain-, Harp-, Brown-dikes) as well as the shared Gardar-Voisey's bay fault zone that led to the emplacement of both the Nain and Gardar magmas in the upper crust.

This study identifies a sequence of plate tectonic and evolutionary processes that favored the generation of highly REE-enriched alkaline silica-undersaturated and oversaturated melts. These processes are consistent with the tectonic framework that produced the alkaline magmatism of the Mesoproterozoic Nain and Gardar provinces, and help constrain exploration models for REE-mineralized intrusive bodies.

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FIGURE CAPTIONS

Fig. 1 An Archean and Paleoproterozoic terrane map of Northeastern Québec and Labrador showing the Nain Plutonic (AMCG) Suite and other Elsonian intrusions (grey), the Napeu Kainiut quartz monzonite (NK), and the locations of the Strange Lake (StL), Flowers River Misery Lake (ML) and Ytterby (Y) 1, 2 and 3 REE-rich intrusions (modified after Emslie et al. 1994 and David et al. 2011)

Fig. 2 A geological map of the Strange Lake pluton, host Napeu Kainiut quartz monzonite and Core Zone gneisses. Also shown are the locations of the samples used for this study. The white dots represent the sampling locations of Kerr (2015) on the Labrador side of the pluton

Fig 3 Chondrite normalized REE-profiles for the bulk rock samples used in this study. The normalization was done using the chondrite data of Sun and McDonough (1989)

Fig. 4 A binary diagram showing bulk rock concentrations of Sm and Nd in the Strange Lake granites (this study and Kerr 2015) and the host Napeu Kainiut quartz monzonite. Concentrations of both elements increase with increasing evolution of the granites

Fig. 5 A binary diagram showing bulk rock and arfvedsonite ¹⁴⁷Sm/¹⁴⁴Nd and ¹⁴³Nd/¹⁴⁴Nd ratios for the Strange Lake granites (this study and Kerr 2015), and the Napeu Kainiut quartz monzonite. The data for the Strange Lake granite define a single linear correlation that is roughly parallel to a linear correlation for the Napeu Kainiut quartz monzonite but at a significantly higher ¹⁴³Nd/¹⁴⁴Nd ratio

Fig 6 Binary diagrams showing a) ɛNd vs Nd concentration and b) ɛNd vs. ¹⁴⁷Sm/¹⁴⁴Nd. The Nd concentration increases with granite evolution reflecting in-situ crystal fractionation by LREE-minerals and is accompanied by a similar increase in the ¹⁴⁷Sm/¹⁴⁴Nd ratio

Fig 7 A plot of model ages (T_{DM}) vs. ϵ Nd (t) for the Strange Lake granites (bulk rock, this study and Kerr (2015), and arfvedsonite) for 147 Sm/ 144 Nd ≤ 0.13 at t = 1.24 Ga, and for the Napeu Kainiut quartz monzonite at t = 1.42 Ga. The line and curve representing CHUR and depleted mantle, respectively, are from De Paolo (1988)

Fig. 8 A diagram comparing the range of δ^{18} O values (Table 4) for the Strange Lake granites and Core Zone gneiss (this study) to corresponding values for global reservoirs. The mantle value of 5.7 ‰ ± 0.2 (Ito et al. 1987) is shown by the red dashed line. The data for the global reservoirs were taken from Onuma et al. (1972); O'Neil (1977); Radain et al. (1981); Harmon and Hoefs (1985); Hoefs (1987); and Wu et al. (2003)

Fig. 9 a) A diagram for the AFC model showing the dependence of the evolutionary path for δ^{18} O on the δ^{18} O value of the contaminant (10 to 18 ‰) and the value of 'r', the ratio between the mass assimilation rate and the fractional crystallization rate (0.1 to 0.8). The range of δ^{18} O values for the Strange Lake granites is best explained by a contaminant with a δ^{18} O value of 11‰ and a 'r' value between 0.3 and 0.4 (see text section 5.6 for further detail); b) A diagram comparing the effect of fractional crystallization 'FC' (Rayleigh fractionation) with the combined effects of fractional crystallization and assimilation 'AFC' on the δ^{18} O evolution of a magma with an initial δ^{18} O value of 6.3 ‰ and a contaminant with a δ^{18} O of 11‰

Fig. 10 A plot of εNd versus Nd concentration showing the exponential increase in melt Nd concentration with decreasing εNd value upon fractional crystallization and assimilation of 5, 10, 15 and 25 % of crustal material. See text section 5.6 for further detail

Fig. 11 A diagram showing the evolving ε Nd and δ^{18} O isotopic composition with assimilation and fractional crystallization of a basaltic starting composition corresponding to the least contaminated Voisey's Bay intrusion (olivine gabbro) (ε Nd = +0.9 and δ^{18} O = +6.3) with a

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contaminant having the composition $\varepsilon Nd = -15$ and $\delta^{18}O = +11$ (see text section 5.6 for references and further explanation). The evolution of the isotopic composition is displayed as a function of melt fraction crystallized (f). The compositional range of the main gneiss types in the vicinity of the Strange Lake pluton and elsewhere in the Core Zone (biotite paragneiss, enderbitic and quartzofeldspathic orthogneiss) is displayed as a grey box for reference

Fig. 12 Sketches illustrating the tectonic processes that affected the Core Zone between 2.2 and 1.24 Ga. a) Initial rifting, b) Subduction, c) Continent-continent collision, and d) Elsonian magmatism. The red star represents the location of the Strange Lake pluton. SCLM = subcontinental lithospheric mantle

Fig. 13 A simplified map showing the tectonic units of northeastern Laurentia. The distance between NE-Canada and SW-Greenland is disproportional. The locations of the Nain Plutonic Suite (Québec-Labrador) and the Gardar alkaline intrusions (SW-Greenland) are shown in grey, the inferred link between the Ketilidian and Makkovik provinces by black dashed lines, and the Gardar-Voisey's bay Ni-Cu-deposit and fault zone connecting both provinces by a red dashed line







doi: ARTN 54 10.1007/s00410-017-1373-x.12/16



Fig. 4













doi: ARTN 54 10.1007/s00410-017-1373-x.12/16





Fig. 12



A Nd- and O-isotope study of the REE-rich peralkaline Strange Lake granite: Implications for Mesoproterozoic A-type magmatism in the Core Zone (NE-Canada)

Contributions to Mineralogy and Petrology

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Tables

Unit	Strange Lake			Napeu Kainiut		
Rock	Hyper-	Hyper-	Quench	Trans-	Quartz	Quartz
type	solvus	solvus	zone	solvus	monzonite	monzonite
сурс	granite S	granite N	granite	granite	inside SL	outside SL
Sample	10032	10040	10036	10039	10118	10216
SiO ₂	69.13	71.16	70.62	71.61	75.26	70.92
Al_2O_3	12.23	11.97	11.33	10.82	11.84	12.69
Fe_2O_3	5.26	4.82	4.90	5.53	2.44	5.19
MnO	0.07	0.10	0.09	0.12	0.03	0.08
MgO	0.03	0.05	0.03	0.03	0.08	0.26
CaO	0.65	0.61	0.60	0.55	0.75	1.68
Na ₂ O	5.06	5.00	5.15	6.56	2.98	3.02
K ₂ O	5.31	4.74	4.47	2.88	5.67	5.05
TiO ₂	0.21	0.27	0.19	0.21	0.21	0.56
Nb_2O_5	0.01	0.05	0.04	0.14	0.004	0.004
P_2O_5	-	0.02	0.01	-	0.01	0.11
F	0.42	0.41	0.44	0.49	0.17	0.07
TREO	0.11	0.21	0.24	0.41	0.09	0.06
LREO	0.06	0.14	0.16	0.27	0.07	0.05
HREO+Y	0.04	0.07	0.08	0.14	0.02	0.01
Total	98.59	99.63	98.36	99.75	99.72	99.95
LOI	0.33	0.49	0.24	0.29	0.43	0.41
AI	1.15	1.12	1.17	1.29	0.93	0.82
Be (ppm)	14	47	51	96	9	4
Zn	210	300	350	520	40	90
Ga	45	40	50	60	24	22
Rb	434	454	512	467	311	188
Sr	12	17	22	13	46	134
Zr	1122	3524	2620	4971	349	640
Sn	31	69	65	103	5	6
Cs	2	6	1	4	3	2
Ba	39	73	53	45	313	1374
Hf	33	83	77	136	11	14
Та	7	20	18	43	2	1
Pb	43	97	129	206	42	33
Th	22	80	74	98	38	18
U	4	15	13	21	8	3

Table 1: Bulk rock major element (wt. %) and trace element (except REE and Y) concentrations of representative samples from the different lithological units sampled in this study.

Unit	Strange Lake				Napeu Kainiut	
Pock	Hyper-	Hyper-	Quench	Trans-	Quartz	Quartz
type	solvus	solvus	zone	solvus	monzonite	monzonite
type	granite S	granite N	granite	granite	inside SL	outside SL
Sample	10032	10040	10036	10039	10118	10216
La (ppm)	108	252	291	493	136	96
Ce	235	540	589	1010	272	193
Pr	28	62	71	120	31	22
Nd	106	221	265	438	113	88
Sm	25	49	59	100	20	16
Eu	2	3	4	6	1	2
Gd	24	45	55	95	17	12
Tb	5	9	11	18	3	2
Dy	33	63	72	116	17	11
Но	7	14	16	25	3	2
Er	23	48	50	75	10	7
Tm	3	8	8	11	2	1
Yb	22	53	50	73	10	6
Lu	3	8	7	10	2	1
Y	245	395	460	773	92	58
LREE	527	1171	1333	2262	591	430
HREE	96	203	213	329	46	30
REE	624	1374	1546	2590	637	460

Table 2: Bulk rock REE and Y concentrations of representative samples from the different lithological units sampled in this study.

Table 3: Sm-Nd concentrations, isotope data and ages for the Strange Lake granitic pluton and the Napeu Kainiut quartz monzonite plus depleted mantle model ages (T_{Dm}).

Трм
<u> </u>
2.02
1.95
1.07
2.35
2.19
2.20
2.66
2.44
1.94
3.18
2.16
2.29
2.18
2.42
1.77
2.72
2.39
2.38
2.69
2.05
2.75
2.04

a) Analytical errors for ¹⁴⁷Sm/¹⁴⁴Nd are estimated to be less than 0.1% (~ 0.0001 to 0.0004), and for ¹⁴³Nd/¹⁴⁴Nd are estimated to be less than 0.002% (~0.00001) at 2 sigma. The calculation of model ages in respect to the depleted mantle was based on the model of Jacobson, 1984.
b) The initial isotope compositions for the Strange Lake granites were calculated using the 1.24 Ma U-Pb age from Miller et al. (1997). The values for the Napeu Kainiut quartz monzonite host rock were calculated assuming an age of 1.42 Ga, the age of the Mistastin batholith (Emslie & Stirling, 1993).

Table 4: Oxygen isotope compositions of quartz separates from the Strange Lake pluton (StL), the Napeu Kainiut quartz monzonite (QM) with location (NK-i = inside; NK-o = outside the Strange Lake pluton) and bulk samples of gneiss with location (GN-i = inside; GN-o = outside the Strange Lake pluton)

Sample	Unit	Rock type	Material	δ ¹⁸ O (‰)
10010-Q	StL	Hypersolvus granite - north	quartz	8.7
10014-Q	StL	Transsolvus granite	quartz	8.8
10016-Q	StL	Hypersolvus granite - south	quartz	8.4
10039-Q	StL	Transsolvus granite	quartz	8.9
204749-Q	StL	Quench zone granite	quartz	8.4
204773-Q	StL	Hypersolvus granite - south	quartz	8.2
46-B-3	StL	Transsolvus granite	quartz	8.6 *
48-A-1	StL	Hypersolvus granite - north	quartz	8.5 *
MBTR-3	StL	Transsolvus granite	quartz	9.1 *
10118-Q	NK-i	Quartz monzonite	quartz	8.9
206133-Q	NK-o	Quartz monzonite	quartz	9.1
204812-В	GN-o	Biotite gneiss banded	bulk	8.6
204824-В	GN-i	Biotite gneiss banded	bulk	9.6
206127-В	GN-i	Garnet-biotite gneiss banded	bulk	6.3

* data from Boily and Williams-Jones (1994)

Table 5: Nd-concentrations (ppm), isotope values and ages of basement rocks plus average depleted mantle model ages (T_{Dm}).

Rock type (n)	Location	\mathbf{Nd}^*	T (Ga)	εNd (t) avg.	Range	Т _{DM} (Ga)	Reference
Churchill Province / Core Zone (Paleoproterozoic)							
Gneiss (granitoid) (2)	near Strange Lake	15	1.24	-12.7	-12.4, -13.0	2.37	Kerr, 2015
Orthogneiss granitoid (10)	Core Zone	-	1.30	-14.6	-11.1 to -18.3	2.70	Emslie et al., 1994
Enderbitic orthogneiss (3)	near Voisey's bay	-	1.32	-4.0	-2.8 to -5.7	-	Amelin et al., 2000
Tasiuyak paragneiss (4)	near Voisey's bay	30	1.32	-9.2	-8.3 to -10.1	-	Amelin et al., 2000
Nain Province (Archean) TTG and felsic gneiss (8) Orthogneiss (5) (Banded) orthogneiss (3) Gneiss Archean (1)	near Kiglapait near Voisey's bay Makkovik Province Gardar Province	15 50 25 36	1.30 1.32 1.72 1.29	-21.9 -14.2 -15.6 -17.8	-14.2 to -28.1 -11.6 to -18.3 -14.8 to -16.5 -17.8	3.20 - 3.10 -	Emslie et al., 1994 Amelin et al., 2000 Kerr & Fryer, 1993 Bartels et al., 2015
Least contaminated mafic magmas							
Ol-gabbro, troctolites (22)	Voisey's bay	-	1.32	-1.2	+0.9 to -2.1	-	Amelin et al., 2000
Ol-bearing mafic dikes (5)	Nain Province	40	1.28	-3.0	-0.2 to -4.6	2.16	Carlson et al., 1993
Mafic dike swarm (12)	Gardar Province	21	1.29	-2.3	-0.5 to -4.4	-	Bartels et al., 2015
Eriksfijord basalt (6)	Gardar Province	18	1.17	+2.1	+2.0 to 2.2	-	Paslick et al., 1993

* Avg. Nd concentrations of bulk rock samples. If no value is reported, the isotopic analysis was done on a mineral fraction.

t = age for the calculated $\epsilon Nd(t)$, which does not necessarily represent the age of the rock.

Table 6: Bulk rock δ^{18} O values of for Archean and Paleoproterozoic rocks in the Churchill and Nain Provinces.

Rock type (n)	Location	δ ¹⁸ O avg. (‰)	δ ¹⁸ O range (‰)	Reference
Churchill Province / Core Zone (Paleop	<u>roterozoic)</u>			
Enderbitic (ortho) gneiss (28)	near Voisey's bay	7.2	6.7 - 8.7	Ripley et al., 2000
Tonalite / biotite gneiss (2)	near Voisey's bay	6.8	6.1, 7.5	Peck et al., 2010
Tasiuyak paragneiss (26)	near Voisey's bay	10.9	8.3 - 16.1	Ripley et al., 2000
Tasiuyak paragneiss (2)	near Voisey's bay	9.5	9.2, 9.7	Peck et al., 2010
Nain Province (Archean)				
Mafic gneiss (14)	near Voisey's bay	7.3	5.7 - 8.5	Ripley et al., 2000
Qtz-diorite gneiss / migmatite (4)	near Voisey's bay	6.4	5.4 - 7.4	Peck et al., 2010
Quartzofeldspathic gneiss (2)	near Voisey's bay	9.6	9.5, 9.6	Ripley et al., 2000
Least contaminated mafic magmas				
Olivine-gabbro, troctolites (106)	Voisey's bay intrusion	6.3	4.6 - 8.2	Ripley et al., 2000
Olivine-bearing mafic dikes (5)	Nain Province	6.8	6.3 - 7.9	Carlson et al., 1993

Appendix

<u> ϵ Nd - δ^{18} O endmember mixing</u>

Eq (A.1)

Because $D_{Nd} \ll 1$:

$$\frac{\varepsilon_m - \varepsilon_m^0}{\varepsilon_a - \varepsilon_a^0} = 1/(1 + \frac{C_m^0 \cdot M_m}{C_a \cdot M_a})$$

with

$\frac{C_m}{C_m^0} \sim F^{-1} \left[1 - \frac{C_a}{C_m^0} \cdot \frac{r}{r-1} \right]$
--

Eq (A.2)

Because D $\delta^{18}O\sim 1$ and $r\neq 1$

$$\delta_m - \delta_0 = (\delta_a - \delta_0 - \frac{\Delta}{r}) \cdot (1 - F^{(-z)})$$

with

$$z = \frac{r}{r-1}$$

Table A1: Parameters used for mixing calculations

	parameter
Source	
c Nd _l	40 ppm
ϵNd_i	+ 0.9
$\delta^{18}O_i$	6.3 ‰
Contaminant	
c Nd _c	30 ppm
ϵNd_c	-15.0
$\delta^{18}O_c$	+11 ‰
rate 'r'	0.35