

## Hf–Nd–Pb isotope evidence from Permian arc rocks for the long-term presence of the Indian–Pacific mantle boundary in the SW Pacific

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### Abstract

Trace element and initial Hf–Nd–Pb isotopic composition of a series of late Paleozoic arc rocks from South Island, New Zealand show evidence for the presence of Indian-type mantle in the Permian SW Pacific. The trace element budget points to a fluid-dominated arc setting such that Pb isotope compositions for both the volcanic and intrusive rocks were controlled by addition of fluids derived from the subducted slab to the mantle wedge. Relatively unradiogenic initial  $^{207}\text{Pb}/^{204}\text{Pb}$  ratios indicate only a negligible contribution of pelagic sediments to the subduction component. Initial  $^{208}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$  co-variations therefore indicate that the subduction component originates from subducted oceanic crust having a Pacific-type composition. In contrast, Hf–Nd isotope correlations, corrected for slab fluid addition, reveal an Indian-type signature for the mantle wedge. Thus, the results indicate contribution of material from both Pacific- and Indian-type mantle sources to the island arc melts. From the source variability in Hf–Nd–Pb isotopes, it is therefore evident that a mantle domain boundary was present beneath the Permian Brook Street arc, similar to the prominent present-day isotope mantle boundary in the Earth's upper mantle, which can be traced along the western Pacific rim. These observations provide strong support that the isotopically defined mantle boundary between Indian and Pacific-type mantle was present in the SW Pacific since at least the late Permian. The existence of this boundary implies that convection cells of the Pacific and Indian mantle reservoirs co-existed in close proximity and yet remained distinct and isolated from each other since at least the late Permian. These results provide strong indirect evidence for the absence of significant chemical exchange between neighboring convecting regimes, at least for the approximate duration of one mantle overturn. Applying these distinct isotope features to the Permian plate tectonic configuration, the subduction polarity of the Brook Street arc was facing westwards, towards the active SE Gondwana margin.

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## 1. Introduction

The upper mantle beneath the Indian Ocean is distinct in its geochemical composition, and can be clearly distinguished from the mantle beneath the Pacific Ocean. The distinction is based on the characteristic isotope compositions of the mid ocean ridge and island arc basalts derived from both mantle reservoirs [1–3]. Indian-type mantle, or sometimes referred to as DUPAL-type mantle (which is the EM1 ocean island basalt end member in the southern Atlantic Ocean, e.g., [4,5]), has long been known to be distinct from Pacific-type mantle in its Sr, Nd and Pb isotope compositions [1,6–9]. The recent advent of multiple collector plasma source mass spectrometry (MC-ICPMS) facilitated Hf isotope measurements for small sample amounts, thus permitting the application of Hf–Nd co-variation diagrams for further distinction between both reservoirs [3,10–12].

Various geophysical and geochemical studies were carried out to determine the nature and origin of this prominent geochemical boundary that is currently located in the Western Pacific [2,13–17]. In general, Indian-type mantle is characterized by more radiogenic Sr isotope compositions and, relative to Pacific-type mantle, by higher  $^{208}\text{Pb}/^{204}\text{Pb}$  for a given  $^{206}\text{Pb}/^{204}\text{Pb}$ . Moreover, Indian-type mantle is characterized by less radiogenic Nd at a given Hf isotope composition when compared to its Pacific counterpart. The observed distinction in Hf–Nd–Sr–Pb isotopes between these two mantle reservoirs is currently interpreted to be the result of the upper Indian mantle domain having been contaminated with continental material [6]. To produce the observed Indian-type isotope signature from a Pacific-type mantle, the contaminating material must have more radiogenic Sr and Pb, and less radiogenic Nd and Hf isotope compositions. The nature of the contamination, however, still remains a matter of debate. Various workers suggested that the radiogenic Sr isotope signature in Indian-type mantle is due to long-term inputs of subducted [6], ~1.5 Ga old sediment [2] into the upper Indian mantle. Other models have invoked multiple plume input [9], delamination of subcontinental lithosphere [18,19], delamination of lower continental crust [20], or mantle wedge depletion by long-term subduction [21] to account for the upper mantle heterogeneity beneath the Indian Ocean.

The age of the Indian mantle domain is not yet well defined. Crawford et al. [22] reported Indian-type isotope signatures in arc rocks in the Central New Hebrides arc that are as old as 60 Ma. A mantle errorchron with a combined model age of  $\sim 250 \pm 50$  m.y. has been suggested for the origin of this feature based on Nd–Sr–Pb isotope systematics [23]. Xu and Castillo [24] reported

isotope correlations with affinities to Indian-type mantle in northern Chinese ophiolites that are as old as 360 Ma. Early Carboniferous Indian-type signatures have also been reported from other Tethyan ophiolites [25].

Along with the geochemical distinction, a geophysical discrimination into two convection cell systems, generally coupled to subduction activity, has been postulated [1]. This convection cell boundary between Indian-type mantle and Pacific-type mantle is presently observed in subduction environments along the western Pacific rim, from the Southern Ocean South of Australia [1] to Kamchatka in the North [12] (Fig. 1). The boundary between the Indian and Pacific mantle domains is always coupled to subduction activity with the only exception at the Australian Antarctic discordance (AAD), south of Australia [1]. The AAD is characterized by an area of depressed seafloor topography across the South-East Indian Ridge, which is thought to be caused by a fossil slab fragment from a palaeo-subduction zone that is trapped at shallow upper mantle levels [1,26–28]. Palaeo-plate reconstructions for the Cretaceous show that the AAD was located at the eastern margin of Gondwana [26]. This is the area in eastern Gondwana that experienced a long history of subduction activity during the Palaeozoic and Mesozoic. From this correlation it seems most likely that the slab fragment underneath the AAD might have been part of the western Panthalassic Ocean arc system.

To evaluate the time-integrated evolution of the isotopic mantle boundary between the Pacific and Indian oceans, we studied a sequence of primitive igneous rocks of the Permian Brook Street Terrane on the South Island of New Zealand. These rocks are well preserved and are overprinted only by prehnite-pumpellyite facies metamorphism. The rock assemblages of the Brook Street Terrane were emplaced in the western Pacific region east of SE Gondwana. The subduction zone, which acted as the potential precursor of the present AAD, was located in a similar palaeo-tectonic position. Hence, the Brook Street Terrane rocks potentially provide an excellent opportunity to assess the possible presence of the two mantle domains in Permian time.

## 2. Geologic background

The Permian Brook Street Terrane is one of several Palaeozoic to Mesozoic crustal blocks recognized in the Eastern Province of New Zealand ([29], and references therein). The Palaeozoic volcanic rocks of the Brook Street arc represent an intra-oceanic island arc chain in the Panthalassic Ocean that developed around ~260 Ma [30], outboard from the margin of SE

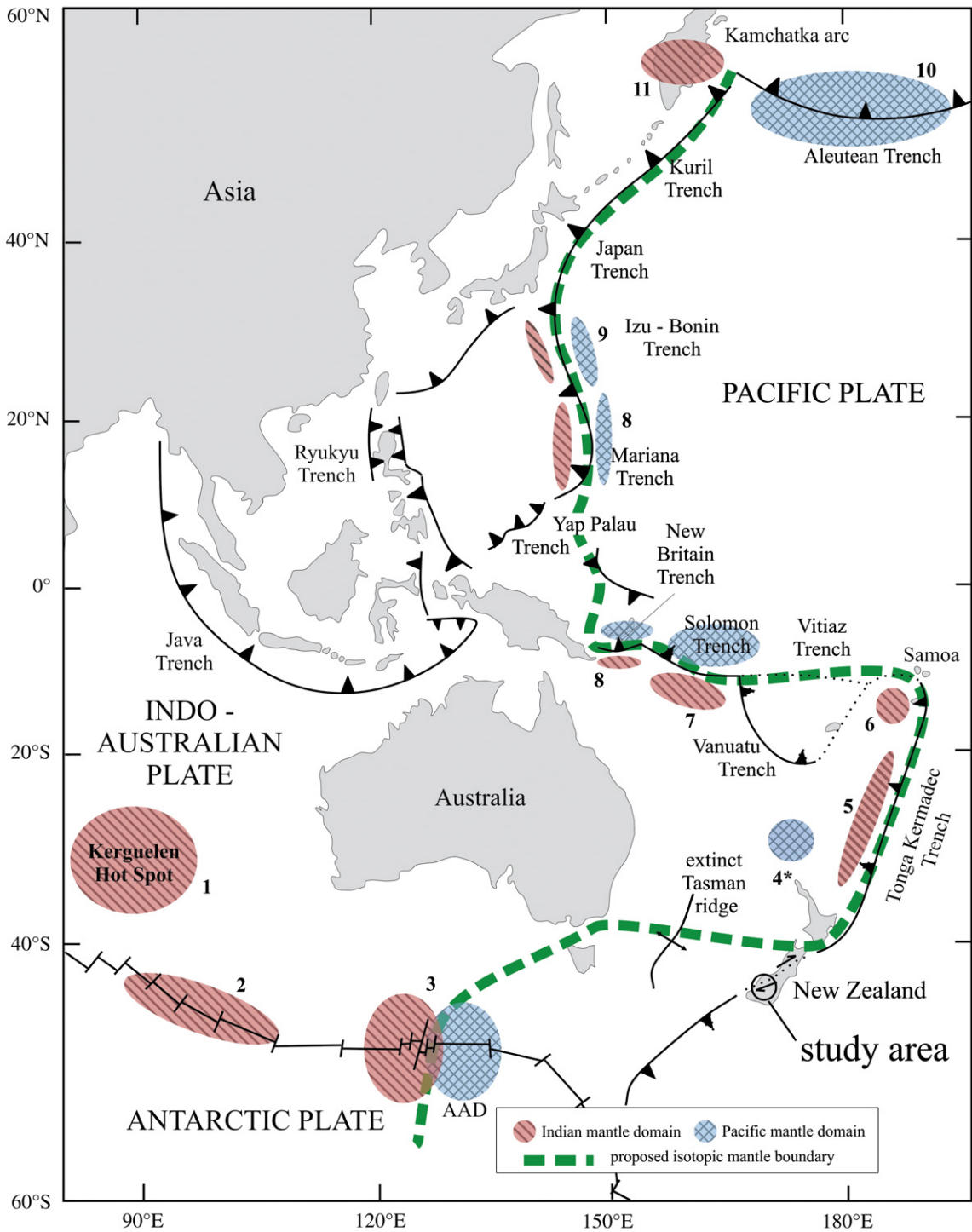


Fig. 1. Plate tectonic overview of the present day western Pacific rim, including the study area. The dashed green line shows the postulated isotopic mantle boundary between Indian- and Pacific-type mantle based on the following studies: 1 — [23]; 2 — [66,67]; 3 — [1]; 4 — [68]; 5 — [22], [69]; 6 — [70]; 7 — [10]; 8 — [71]; 9 — [3]; 10 — [12]; 11 — [72].

Gondwana and accreted to this supercontinent in the early Triassic [31]. Subsequent plate movements in the late Cretaceous to early Tertiary, caused by the now-extinct Tasman spreading ridge, separated the continental fragment of New Zealand, including Brook Street arc rocks, from Australia to its present isolated location in the western Pacific [32]. The sample suite from the Brook Street Terrane investigated in this study is exposed in the southern Takitimu Mountains and in the adjacent Longwood Range on the southern South Island of New Zealand (Fig. 2).

The mafic volcanic rocks from the Takitimu Mountains are predominantly tholeiitic [33]. The rock suite comprises submarine extrusive and shallow intrusive volcanic rocks, interbedded in a voluminous, ca. 16-km thick sequence of marine lithic sandstone, all overprinted by prehnite-pumpellyite facies metamorphism [34]. In contrast, Longwood Range samples comprise plutonic rocks with tonalite–trondjemite–granodiorite (TTG) affinities. These plutonic rocks were emplaced from 260 Ma to ~230 Ma, whereas the youngest rocks intruded coevally with or shortly after accretion of the arc front to Gondwana [31]. In contrast to the currently accepted petrogenetic model for most TTG rocks, which supports an origin of TTG rocks from melting of young, subducted lithosphere [35], the suite from the Longwood Range formed dominantly by re-melting of underplated crust [31,33].

### 3. Results

#### 3.1. Petrography of volcanic rocks

The volcanic flows and crosscutting dikes have porphyritic texture with pyroxene and amphibole phenocrysts in a fine groundmass that is mainly composed of

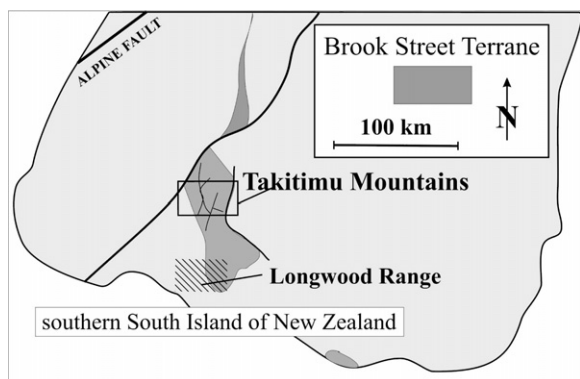


Fig. 2. Map of the southern South Island, New Zealand, showing locations of the sample areas.

feldspar and accessory magnetite. Two major groups of igneous rocks can be distinguished based on their petrography. The first group is dominated by primary magmatic clinopyroxene (cpx, 5–15 vol.%), feldspar (20–70 vol.%) and magnetite (1–5 vol.%). This group occurs as both volcanic flows and shallow intrusive sills. Amphibole-phyric cross cutting dikes (5–30 vol.% amphibole) constitute the second group. In these rocks, some amphiboles show magmatic rims of augite and vice versa. Green celadonite and calcite can be observed as secondary metamorphic phases along fracture zones. Prehnite and pumpellyite are much less abundant than zeolite. The occurrences of all secondary minerals seem to be restricted to major fracture zones, micro-scale fractures and/or vugs. A detailed petrographic description, and associated age spectra of Longwood Range samples are given in Mortimer et al. [31].

#### 3.2. Major and trace element composition

##### 3.2.1. Takitimu Mountains samples

Major and trace element data and a description of all analytical procedures are available as online supporting material.

Volcanic rocks from the Takitimu Mountains cover a range from basaltic to andesitic compositions with total SiO<sub>2</sub> contents varying from 48.9 to 61.8 wt.%, with no significant gap in the range of SiO<sub>2</sub> contents. Total alkalis increase slightly with increasing silica contents and overall low K<sub>2</sub>O contents of 0.2 to 2.8 wt.% indicate affinities to typical low-K volcanic arc rock series. Although this is apparently consistent with a primary magmatic origin, alkali element contents were most likely disturbed by low-grade metamorphic mobilization. Two samples show significantly elevated [Na<sub>2</sub>O + K<sub>2</sub>O] contents, which most likely are due to secondary enrichment caused by vug fillings with zeolites. MgO contents of the lavas vary from 2.0 to 9.0 wt.%. Contents of TiO<sub>2</sub> increase only slightly with decreasing MgO, probably due to early magnetite fractionation.

Volcanic rocks of the Takitimu Mountains from clinopyroxene and amphibole suites show similar FeO contents at MgO > ~5.8 wt.%, whereas at lower MgO values, FeO contents in the amphibole suite are lower than in the clinopyroxene suite. This suggests increasingly hydrous conditions and enhanced magnetite fractionation during differentiation of the amphibole suite. Increasing Al<sub>2</sub>O<sub>3</sub> values in both suites with decreasing MgO indicate no significant fractionation of plagioclase, but are consistent with clinopyroxene and/or amphibole fractionation. Nickel contents of ≤ 120 ppm in both suites point to an early fractionation of olivine at shallow to intermediate

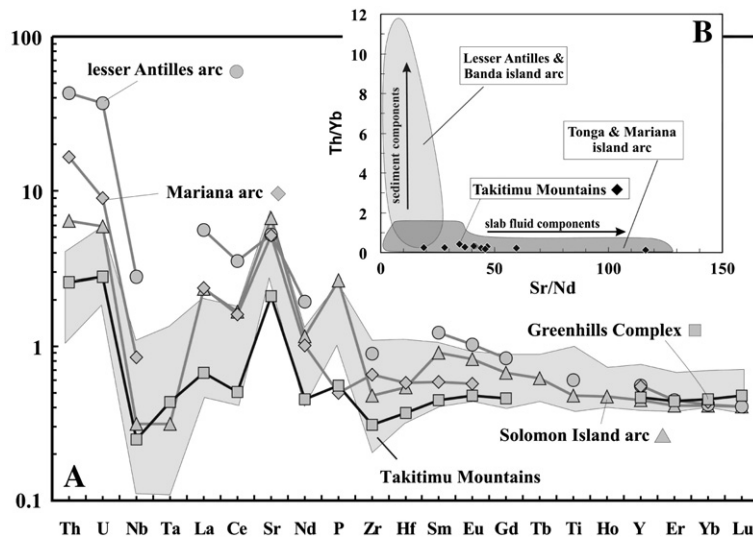


Fig. 3. Trace element abundances in rocks of the Takitimu Mountains volcanic suite in comparison to present-day and Permian arc rocks. (A) N-MORB-normalized incompatible trace element diagram. Trace element patterns from intrusive Brook Street Terrane rocks of the Greenhills complex are shown for comparison [73]. The overlap between the two suites supports that element abundances reflect primary magmatic signatures. Lesser Antilles data are from [74]; Mariana data from [3]; [11], Solomon Islands data are taken from [10], (B) Plot of Th/Yb versus Sr/Nd in comparison to presently active arcs [11], suggesting an intra-oceanic arc setting for the Takitimu samples, where significant continent-derived components are absent.

crustal levels. Most mobile, large-ion lithophile elements (LILE, e.g., K, Cs, Rb, Ba) show rather large scatter among all samples compared to immobile elements such as REE (rare earth elements) or HFSE (high field strength elements). The LILE were most likely mobilized during the zeolite to prehnite-pumpellyite facies overprint [34], and are thus excluded from further interpretation of primary magmatic processes. Abundances of immobile trace elements (i.e., Th, HFSE, REE) in the volcanic rocks are consistent with those in modern island arc settings

(Fig. 3A) and are thus apparently not affected by metamorphism. Characteristic Nb–Ta and Zr–Hf depletions relative to REE, and consistent Sr enrichment (Fig. 3A) in trace element patterns, indicate a subduction zone setting [36], where the depleted mantle wedge was enriched by LILE-rich subduction components. Th/Yb are consistently low (< 1) when compared to modern arc systems, which is an argument for an intra oceanic arc environment with very low terrigenous sedimentary input. Elevated Sr/Nd indicate addition of subduction fluids (Fig. 3B).

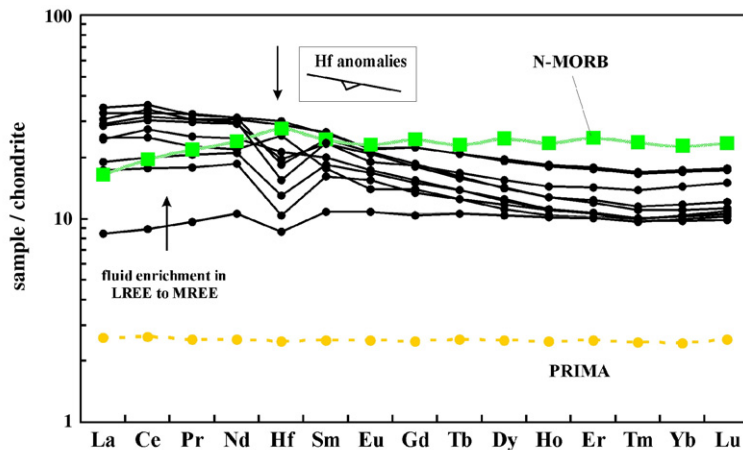


Fig. 4. Extended REE patterns (including Hf) of Takitimu Mountains volcanic rocks normalized to chondritic values [75]; negative Hf anomalies indicate the addition of light REE to the mantle wedge by subduction components. N-MORB and PRIMA after Hofmann [76].

Chondrite-normalized REE patterns show only slightly enriched light REE (Fig. 4).

### 3.2.2. Longwood Range samples

Plutonic samples from the Longwood Range suite cover a wide range in SiO<sub>2</sub> contents from 50 to 70 wt.%, and belong to the tonalite–trondhjemite–granodiorite association [31], as indicated by high Sr/Y and elevated (La/Yb)<sub>N</sub>. The major and trace element chemistry of these rocks is discussed in detail elsewhere [31]. High Sr/Y (>30) as observed in these rocks are often associated with slab melting. A slab melting origin, however disagrees with very low MgO contents (<1 wt. %) even in primitive rocks, which is not typical for slab melts [37].

### 3.3. Hf, Nd, and Pb isotopes

No significant change in the Hf–Nd, and Pb isotope compositions of the rocks (supporting online material) is observed with varying degrees of fractionation as indicated by the ranges in MgO and SiO<sub>2</sub> contents. Volcanic samples from the Takitimu Mountains show initial Hf and Nd isotope values ranging from  $\epsilon_{\text{Hf}260} + 12.0$  to  $+13.4$ , and  $\epsilon_{\text{Nd}260} + 7.4$  to  $+9.7$ , samples from the Longwood Range vary from  $\epsilon_{\text{Hf}260} + 10.4$  to  $+11.4$ , and  $\epsilon_{\text{Nd}260} + 4.6$  to  $+7.8$ . Plutonic samples from the Longwood Range have slightly lower  $\epsilon_{\text{Nd}(t)}$  and  $\epsilon_{\text{Hf}(t)}$  values compared to their eruptive counterparts from the Takitimu region. The youngest sample of the Longwood Range has a significantly lower initial Hf–Nd isotope composition tending towards compositions of continental crust.

The evolution of the Hf–Nd isotope relationship in Indian-type rocks does not necessarily correlate with the evolution of Pb isotopes in Indian-type mantle, because of different behaviors of these elements during fluid transport- and magmatic processes. Several processes can be the reason for the trace element and isotope characteristics of Indian-type mantle, and correlations of Hf–Pb and Nd–Pb isotopes in Permian time can be different to co-variations in present day reservoirs, and thus lead to misinterpretations. We therefore have restricted the evaluation of mantle domain affinities to Hf–Nd investigations, and internal Pb isotope co-variations. In Fig. 5, the initial Hf–Nd isotope compositions of the magmatic rocks from the Brook Street Terrane are compared with those of the Indian- and Pacific-type mantle reservoirs at 260 Ma, i.e., the average estimated emplacement age of the magmatic rocks from the Brook Street arc. All samples from the Takitimu Mountains plot close to the discrimination line

between the Indian and Pacific mantle domains. Two samples from the Longwood Range show Indian-type mantle affinities, two samples plot within the range of Takitimu rocks, one sample has significantly lower Hf–Nd compositions shifted towards the OIB-continental crust field. In general the Longwood samples display a trend from Indian-type signatures towards Pacific-type signatures.

Leached feldspar separates from some of the Takitimu volcanic rocks were analyzed for their Pb isotopic compositions (Fig. 6, online supporting material). Because feldspar contains almost no U, the measured present-day Pb isotope composition of feldspars is virtually identical to the Pb isotope composition at the time of crystallization. [38,39]. The samples show a variation in  $^{208}\text{Pb}/^{204}\text{Pb}$  from 37.73 to 38.36, in  $^{207}\text{Pb}/^{204}\text{Pb}$  from 15.50 to 15.64 and in  $^{206}\text{Pb}/^{204}\text{Pb}$  from 18.03 to 18.70. The Pb isotope ratios in most analyzed samples overlap with those of MOR basalts when compared to age-corrected present day reservoirs in plots of  $^{207}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$  (Fig. 6). Two samples from the Longwood Range and one from the Takitimu

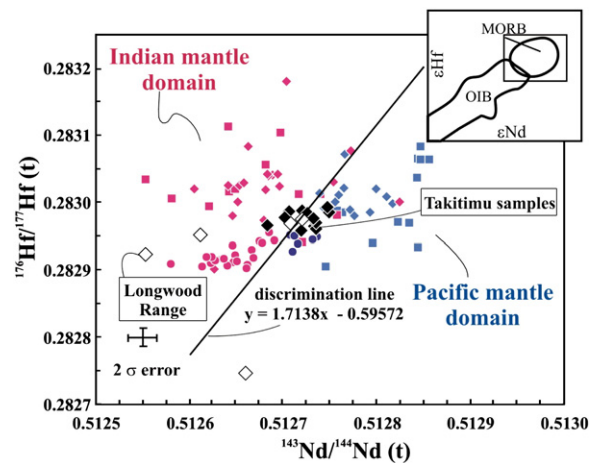


Fig. 5. Initial isotope ratios shown in age corrected Hf–Nd isotope space (260 Ma). The small inset displays the global MORB–OIB array; the MORB field is enlarged. The age corrected discrimination line was taken from [21]. Takitimu rocks are shown as black diamonds, Longwood samples as open diamonds; red symbols represent Indian-type mantle signatures, blue symbols Pacific-type signatures. Squares are taken from [77] and [3]; diamonds from [78]; circles from [21]. Initial Nd and Hf isotopes were calculated for  $t=260$  Ma with a  $\lambda$  ( $^{176}\text{Lu}$ ) of  $1.865 \times 10^{-11}$  [79] and a  $\lambda$  ( $^{147}\text{Sm}$ ) of  $6.543 \times 10^{-12}$  [80]. Chondritic  $^{176}\text{Hf}/^{177}\text{Hf}$  values of 0.282772 and  $^{176}\text{Lu}/^{177}\text{Hf}$  values of 0.0332 [54] were used. Hafnium and Nd age corrections for MORB fields were calculated using the DM values of 0.0381 for  $^{176}\text{Lu}/^{177}\text{Hf}$  [81] and 0.214 for  $^{147}\text{Sm}/^{144}\text{Nd}$  [55]. Note that the Indian mantle potentially has a lower Sm/Nd and Lu/Hf. Using such lower ratios results in a shift of Brook Street Terrane values towards the Indian mantle reservoir.

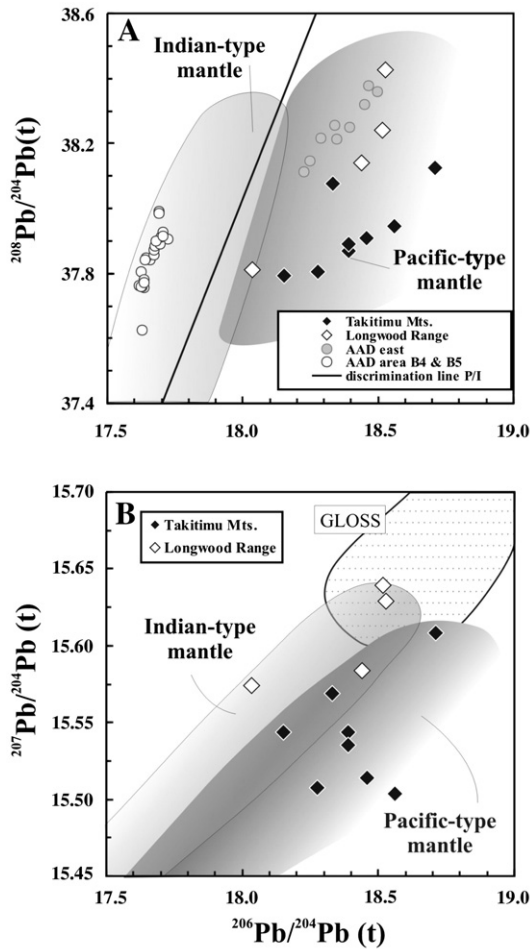


Fig. 6. Lead isotope compositions of leached feldspars from the Longwood Range and the Takitimu Mountains. AAD samples for comparison and discrimination line in figure A are taken from [21]; global subducting sediment (GLOSS) in panel B is taken from [36] and [82]. All present day values are back calculated to  $t=260$  Ma using a  $\mu$  value of 9 and the present day  $^{238}\text{U}/^{235}\text{U}=137.88$ .

Mountains are slightly shifted in their  $^{207}\text{Pb}/^{204}\text{Pb}$  towards the age corrected field of global subducting sediment (GLOSS), most likely indicating some contamination with subducted sediment.

As it is the case for present day arc rocks, samples from both mantle domains overlap in  $^{207}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$  isotope space. In contrast, present day arc rocks show a sharp distinction between Indian vs. Pacific-type mantle in  $^{208}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$  isotope space. In a plot of  $^{208}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$ , samples from both Brook Street Terrane localities indicate a Pacific-type mantle origin for Pb. Hafnium–Nd correlations from Longwood Range TTGs and Pb isotopes indicate the contribution of both Indian- and Pacific-type mantle sources to the magmas. This

discrepancy reflects the different behavior of Pb and Hf–Nd in subduction zone systems.

## 4. Discussion

### 4.1. Petrogenesis and the role of subduction components

Distinguishing between Indian vs. Pacific isotope signatures in arc related rocks is not straightforward because subduction components can have strong and variable influence on the source of the melts in the mantle wedge [3,11]. Therefore, several geochemical criteria were applied to unravel the magmatic evolution of the Brook Street rocks, and the initial isotope signatures of subduction components and the mantle wedge.

#### 4.1.1. Takitimu Mountains suite

A co-genetic origin of the clinopyroxene- and amphibole-dominated suites is indicated by (i) similar compatible and incompatible trace element abundances at similar MgO contents in the most primitive rocks ( $>5.8$  wt.% MgO), (ii) the overall parallel trace element patterns (Fig. 3) for all samples, and (iii) by the overlapping initial  $\epsilon_{\text{Nd}}$  and initial  $\epsilon_{\text{Hf}}$  values. No significant Eu anomalies can be observed in the Takitimu rocks (Fig. 4), confirming major element constraints that plagioclase fractionation was insignificant. Similar to present-day subduction-related rocks, the REE patterns show slightly enriched light REE relative to heavy REE. This REE fractionation reflects preferential partitioning of light REE in the subduction components, whereas the heavy REE are preferentially retained in the slab (e.g., [40–42]). The lower HREE contents compared to N-MORB are a typical feature of subduction rocks, reflecting comparatively higher degrees of melting (e.g., [43]).

In contrast to LREE, Nb and Zr are both immobile during slab dehydration. Zr/Nb vs. La/Yb co-variations can therefore help to assess the degree of mantle wedge depletion relative to that of a typical N-MORB source mantle, and/or variations in the degrees of melting. Typically elevated Zr/Nb in arc rocks are consistent with the model that the mantle beneath presently active arcs is generally more depleted than the MORB mantle source, because a longer history of melt extraction has already depleted the mantle wedge [41].

The enrichment of light REE (elevated La/Yb) in the Takitimu samples is the consequence of their preferential transport by subduction components to the mantle wedge [42]. Hafnium typically displays low solubility in fluids whereas it can show significant mobility in slab melts

[12]. The addition of fluids will therefore mostly affect the Nd budget, while slab melts significantly affect the budget of both Hf and Nd. Trace element patterns of Takitimu Mountain rocks agree in general with typical present day arcs that are fluid dominated, and the presence of slab melts in the sub-arc mantle would be indicated by elevated and much more variable Nb–Ta contents and lower Zr/Nb than in MORB. This feature is not observed, suggesting that the sources of the Takitimu samples do not contain a slab melt component, but were dominated by fluid enrichment. This model is corroborated by high Sr/Nd and low Th/Yb (<1) (Fig. 3B). Present day arc settings, where large amounts of sediments are subducted typically show Th/Yb ratios  $\geq 2$ , whereas fluid dominated arc environments have Th/Yb < 1 (e.g., [11]). Compared to N-MORB (Fig. 7B), the Takitimu lavas show slightly elevated Th contents and similar Nb contents, suggesting that some Th was added to the mantle source. Following Pearce and Peate [36], the mobility of Th in slab fluid is relatively low, but the degree of mobilization depends on fluid composition and pressure [42]. Following Fig. 6B, the relative contribution of Th in Takitimu lavas from slab-derived fluids is  $\sim 80$  to 90% of the total Th content. All these trace element features combined favor the predominance of fluid-like subduction components in the source region of the melts.

#### 4.1.2. Longwood Range suite

Tonalite–trondhjemite–granodiorites, as the Pourakino Trondhjemite [31] from the nearby Longwood Range are abundant in Archean settings and are generally interpreted to result from melting of subducted oceanic crust [44–46]. In Phanerozoic subduction regimes, the occurrence of TTG type suites, or adakites as eruptive analogues, is restricted to regions where either (i) young and hot oceanic lithosphere is subducted [35] or (ii) subducted oceanic lithosphere is melted along the edges of subducting plates [47]. However, an alternative mechanism for producing TTG-type geochemical signatures is re-melting of underplated mafic material in a thickened arc crust where residual garnet and, to a lesser extent amphibole and rutile, are stable [37,44,48]. Since the lower oceanic crust in a thickened arc has a similar mineralogical composition to subducted oceanic crust, the melts also display TTG-type signatures, including high  $(La/Yb)_N$  and elevated Sr/Y > 30. Given the occurrence of TTG-type rocks with Sr/Y > 30 in the Longwood Range, it is possible that either slab melting or remelting of the arc root were the important processes. In Fig. 7a, the presence of residual garnet in the melt residuum is indicated by depleted HREE Nb–Ta and Zr–Hf.

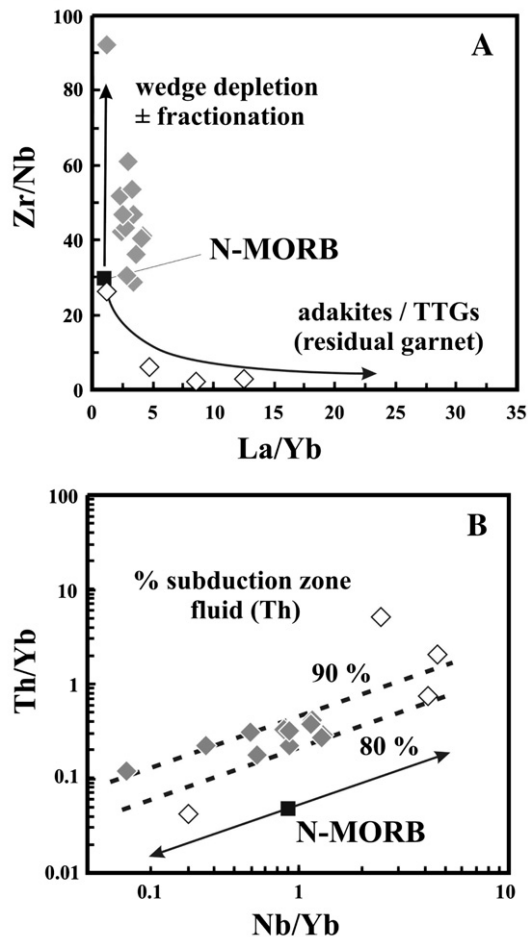


Fig. 7. Plots of trace elements following [36] and [83] to discriminate mantle wedge depletion, the role of slab melts for Brook Street rocks and element mobility. Takitimu samples are grey diamonds, Longwood Range, white diamonds. (A) Plot of Zr/Nb vs. La/Yb, illustrating the absence of slab melt components in Takitimu volcanic rocks, whereas patterns of the Longwood Range rocks indicate the presence of residual garnet in their source, typical for residual eclogite or garnet amphibolite. (D) Plot of Th/Yb vs. Nb/Yb, showing that ca. 80–90% of Th enrichment could have been derived from subduction components. Note the low Th/Yb of <1 that are typical for arc systems where subducted sediments are chemically insignificant.

To distinguish between slab melting or lower crustal melting, we use the MgO content of the samples which is a sensitive tracer for chromatographic processes during mantle percolation. Experimentally derived compositions of pristine melts from oceanic crust have consistently lower MgO contents than 2 wt.% [37] in contrast to typical adakites or TTGs that typically display MgO contents of far more than  $\sim 4$  wt.% (e.g. [49]). These higher MgO concentrations are the result of melt-peridotite interaction as melts migrate through the mantle wedge [48,50–52]. All reported TTGs from the

Longwood Range have low MgO concentrations of <1 wt.% [31,53], implying that Longwood Range TTGs originated from re-melting of slab-like, underplated arc crust.

#### 4.2. Uncertainties in the isotopic discrimination of mantle domains

When tracing mantle domain compositions back in time, significant sources of uncertainty are the assumed parent–daughter ratios that are used to correct the isotope compositions of the present day mantle reservoirs back in time. Because the sources of mantle derived rocks beneath mid ocean ridges and intra-oceanic arc settings are similar, average depleted mantle values for Lu/Hf [54] and Sm/Nd [55] were used to calculate the Permian values. It is likely that Indian-type mantle has slightly lower Lu/Hf and Sm/Nd than average N-MORB, as inferred from its less radiogenic present day Hf–Nd isotopic compositions. Using lower parent–daughter ratios to calculate the Indian mantle isotope reservoir back to the Permian results in Hf–Nd isotope compositions that are more radiogenic, which shifts the initial isotope signatures of the Brook Street Terrane rocks towards Indian-type mantle signatures. Because the elemental budget of the Indian mantle domain in the Permian is not known, a conservative time correction model with uniform values for both reservoirs was chosen.

A second possible source of error for the calculated initial isotope ratios is the uncertainty on the absolute age. However, for Permian ages, the uncertainty of the decay constants and an uncertainty of  $\pm 20$  Myrs on the absolute age have no significant effect on the calculated initial Hf–Nd isotope compositions, because the effect of analytical uncertainties is the dominant parameter.

An additional source of error in distinguishing between the two mantle domains is the slope of the discrimination line and the compositional overlap between the two domains. The mantle domain discrimination line defined at the AAD [21] is slightly steeper in Hf–Nd space than that from western Pacific arc systems [3]. This discrepancy might reflect local variations in the Hf–Nd isotope signatures of both mantle domains along the western Pacific boundary (Fig. 1). However, the mantle domain discrimination presented by Kempton et al. [21] along the AAD south of Australia enables a much sharper separation than the discrimination line by Pearce and co-workers [3] defined further north in the Pacific. This is simply because a smaller data set was used by Pearce et al. [3]. Another argument for the use of the discrimination line of [21] is that the Cretaceous

position of the AAD [26] is similar to that of the Brook Street Terrane in the Cretaceous accretionary complex of SE Gondwana, minimizing the effects of any local variations along the mantle boundary. Therefore, the age corrected AAD discrimination line [21] was chosen in this study. The compositional overlap between the two reservoirs in Hf–Nd isotope space is estimated to be approximately in the range of the external reproducibility of most analyses, i.e.,  $\pm 0.5$   $\epsilon$ -units.

#### 4.3. Hf–Nd–Pb isotope evidence for contrasting mantle domain signatures

The different elemental properties of Hf, Nd, and Pb provide a unique opportunity to trace different components in the sources of island arc rocks. We use  $^{208}\text{Pb}/^{204}\text{Pb}$  and  $^{207}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$  co-variations to investigate isotope signatures from subduction fluids because of the high mobility of Pb. In contrast, Hf–Nd isotopes are useful tracers for mantle wedge compositions. This is because Hf, and to some extent Nd, behave as ‘conservative elements’ in fluid controlled subduction zone processes [36]. However, recent studies [11,56,57] demonstrate that Hf can be mobile in subduction settings if significant amounts of melts from pelagic or volcanoclastic sediment are involved. Based on observations from Kamchatka arc rocks, a fluid dominated regime, Münker et al. [12] have confirmed the conservative element behavior of Hf in the absence of large amounts of sediment derived components.

Based on the trace element characteristics (Th/Yb and Sr/Nd, Fig. 3b) and the measured Pb isotope compositions (i.e., low  $^{207}\text{Pb}/^{204}\text{Pb}$ ), it is evident that the petrogenesis of the Brook Street rocks was dominated by fluids derived from subducted oceanic crust rather than by sediment derived melts. The Hf isotope compositions of these rocks can thus be considered to represent that of the underlying mantle wedge. In contrast to Hf, recent studies have demonstrated that Nd can be partially mobilized in subduction settings [3], which results in decoupling of Hf and Nd isotopes that are usually well correlated. A comparison of measured Hf–Nd isotope signatures with the degree of Nd enrichment relative to the more immobile Hf (e.g., [3]) can help to assess the degree of Nd mobility. This approach is crucial for tracing original mantle wedge signatures prior to the overprint by subduction components. An evaluation of Hf–Nd isotope signatures in the Takitimu arc rocks therefore needs to take into account the possible addition of slab-derived Nd to the mantle wedge.

In contrast to Hf and Nd, the geochemical budget of Pb in subduction-related rocks is almost entirely controlled

by slab components (e.g., [58]) as outlined above. The  $^{208}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$  isotope signatures of the Takitimu samples suggest a Pacific-type origin for Brook Street Terrane rocks, indicating that the subducted slab had a Pacific-type mantle composition. In Hf–Nd isotope space, however, the Takitimu Mountain volcanic rocks plot on the discrimination line between Indian-type and Pacific-type mantle, so that at a first view, Hf–Nd isotopes alone cannot be used to distinguish between these two mantle domains. A correction is required for the possible addition of Nd that, similar to Pb, originates from the subducting Pacific plate. Such a correction would shift the initial composition of the mantle wedge field towards Indian-type mantle.

Because the Longwood Range rocks are lower crustal melts that originate from the arc root, these melts can be considered to carry predominantly the Hf–Nd isotope signature of the mantle wedge and the plate above it, which are isotopically indistinguishable. The Hf–Nd isotope compositions of two of these rocks plot into the Indian-type mantle field with a trend towards the other Brook Street rocks. The Pacific-type Pb isotope characteristics of the Longwood samples is in marked contrast to the Indian-type signatures observed for Hf–Nd isotopes, similar as observed for the Takitimu rocks. Likewise, the Pacific-type Pb isotope signatures may be due to subduction zone fluids that were added to the mantle wedge beneath the arc root.

The youngest sample from this suite, with an age of ~230 Ma, displays significantly lower Hf–Nd compositions. Following previous studies (e.g., [31]) this shift most likely reflects crustal contamination during the latest stage of the arc evolution and accretion to the Gondwana margin, hence, this sample is excluded from the interpretation.

To clearly identify the pristine Hf–Nd isotope characteristics of the mantle wedge sources of both rock suites, any Nd added by a fluid component needs to be subtracted. Pearce et al. [3] suggested an approach to assess the pristine initial Hf–Nd isotope composition of the sub-arc mantle wedge using the relationship between  $\Delta\text{Nd}$  and  $\Delta\epsilon\text{Nd}_{\text{P/I}}$  ( $\text{P/I} = \text{Pacific/Indian}$ ).  $\Delta\text{Nd}$  is defined as the enrichment of Nd relative to Hf in an extended REE plot (Fig. 4). Positive  $\Delta\text{Nd}$  values are caused by addition of a fluid (low Hf/Nd) and/or a sediment melt (low Hf/Nd) to the sub-arc mantle wedge. The second parameter,  $\Delta\epsilon\text{Nd}_{\text{P/I}}$ , is defined as the deviation from the discrimination line between Indian-type and Pacific-type mantle signatures in Hf–Nd isotope space (as shown in Fig. 5). Positive values indicate an Indian-, and negative values a Pacific-type mantle affinity. Assuming Indian type mantle in the wedge, any addition of

subduction components will shift the rock compositions towards positive  $\Delta\text{Nd}$  values.

In a plot of  $\Delta\epsilon\text{Nd}_{\text{P/I}}$  vs.  $\Delta\text{Nd}$ , Takitimu Mountain samples (Fig. 8A) plot at  $\Delta\epsilon\text{Nd}_{\text{P/I}}$  values close to 0, i.e., close to the mantle domain discrimination line shown in Fig. 5. Vectors in Fig. 8A indicate the effects of contributions of various, isotopically distinct subduction components on  $\Delta\epsilon\text{Nd}$ , [3]. It is evident, that all Takitimu rocks were overprinted by subduction components as indicated by elevated  $\Delta\text{Nd}$ . Based on the Indian-type affinities of the Longwood Range rocks, an initially Indian-type composition was inferred above for the mantle wedge beneath the Brook Street rocks. Fig. 8A therefore shows that the Hf–Nd isotope composition of the Takitimu samples is generally consistent with contamination of an Indian-type mantle source that was overprinted by subduction components originating from Pacific-type oceanic crust. Such a Pacific-type signature for the subduction component is also consistent with the  $^{208}\text{Pb}/^{204}\text{Pb}$  vs.  $^{206}\text{Pb}/^{204}\text{Pb}$  co-variations. Noteworthy is that addition of subducted sediment would cause the opposite effect. The observed Hf–Nd isotope co-variation is consistent with an Indian-type signature of the mantle wedge, in agreement with inferences from the Longwood Range rocks. In Fig. 8B the effect of contamination by Pacific type subduction components is illustrated in Hf–Nd isotope space.

#### 4.4. Paleogeographic implications

During Permian time there existed two distinct isotope reservoirs along the Brook Street arc, i.e. the subducting Pacific plate and the Indian-type mantle wedge that are distinguishable by their contrasting Hf–Nd–Pb isotope signatures. Due to these distinct isotopic features of the two converging plates, it is possible to reconstruct the subduction polarity in Permian time. Based on the fact that the Brook Street arc was emplaced outboard from the SE Gondwana margin, the overprint of the mantle wedge by Pacific-type signatures implies a subduction polarity that was facing towards the Gondwana continent. This model is in general agreement with previously proposed models for the eastern Gondwana margin (e.g., [59]). Because the convergent plate boundary at the Brook Street island arc was located outboard from the Gondwana continent, but was accreted ~15 m.y. later to the active continental margin [31], it appears likely that a second subduction zone was present in the hinterland of the Brook Street arc. In any case, an oceanic back-arc basin must have been present between the studied arc front and Gondwana. This oceanic back-arc basin was subducted in the late Permian to early Triassic along the

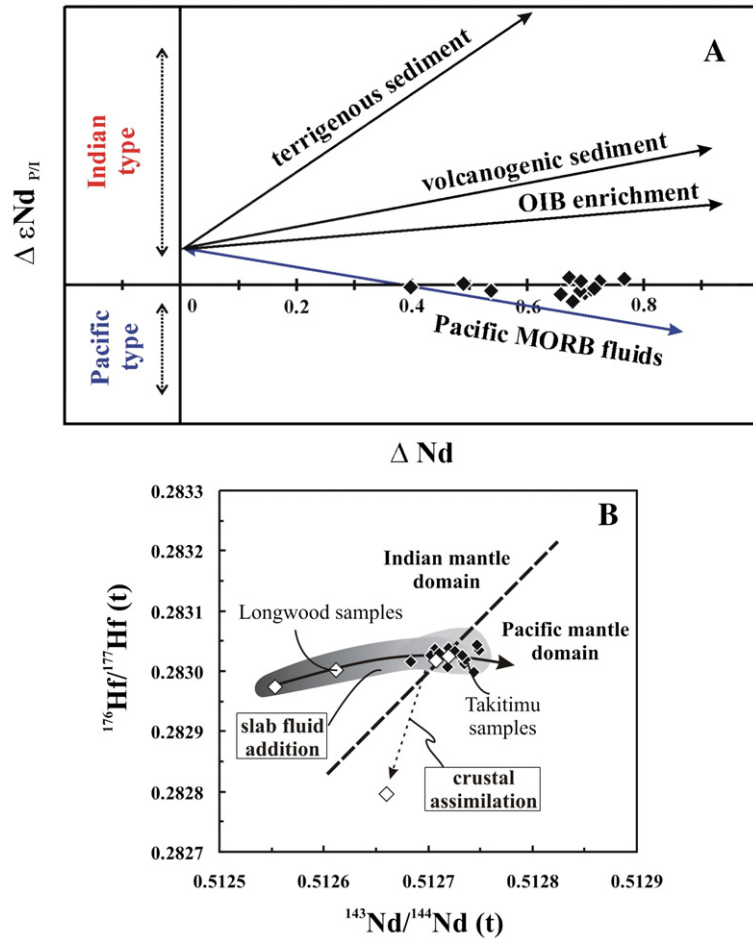


Fig. 8. Hf–Nd systematics in Brook Street Terrane rocks. (A)  $\Delta\epsilon_{NdP/I}$  versus  $\Delta Nd$  projection after Pearce et al. 1999.  $\Delta Nd$  was calculated using equations in Pearce et al. [3];  $\Delta\epsilon_{NdP/I}$  values are determined with the equation:  $\Delta\epsilon_{Nd} = 0.581 * \epsilon_{Hf} - \epsilon_{Nd}$ .  $\Delta Nd$  values indicate the addition of a slab component; positive  $\Delta\epsilon_{NdP/I}$  values indicate Indian type mantle signatures. Arrows indicate typical mixing trends generated by the addition of various subduction components. (B) Age corrected Hf vs. Nd isotope compositions, illustrating the isotopic evolution of Brook Street terrane rocks.

active Gondwana margin until the Brook Street arc chain was accreted to the margin of the Gondwana continent.

The proposed evolution of the mantle domain boundary in the SW Pacific is schematically shown in Fig. 9. After the Brook Street arc was accreted to the Gondwana margin in the Triassic, the location of the mantle domain boundary remained fixed along the convergent margin of south-eastern Gondwana. At this position, the location of the mantle domain boundary was fixed until the Cretaceous when subduction ceased. Cretaceous plate motions were initiated by the break up of central Gondwana. In the course of the continental break up, Australia was separated from Antarctica by the South East Indian Ridge and the continental fragment of New Zealand was separated from Gondwana by the opening of the Tasman Sea along the Tasman Ridge. Following Gurnis et al. [26], the plate motions forced the

Australian continent to drift across the mantle domain boundary. At present, the boundary is located beneath the AAD, south of Australia. During the Cretaceous period of rifting, a fossil slab of subducted oceanic crust was isolated at shallow upper mantle level. Such a stagnated slab fragment may have caused the thermal anomaly, a so-called cold spot [26–28], which is presently linked to the surface depression at the AAD. Gurnis et al. [26] reconstructed the path of this depression and, hence, the path of the associated mantle domain boundary starting from the Cretaceous when it was located underneath the Australian continent to its present position.

#### 4.5. Stable convection since the Permian?

The observation that an isotopically distinct mantle domain boundary in the south-eastern Pacific region

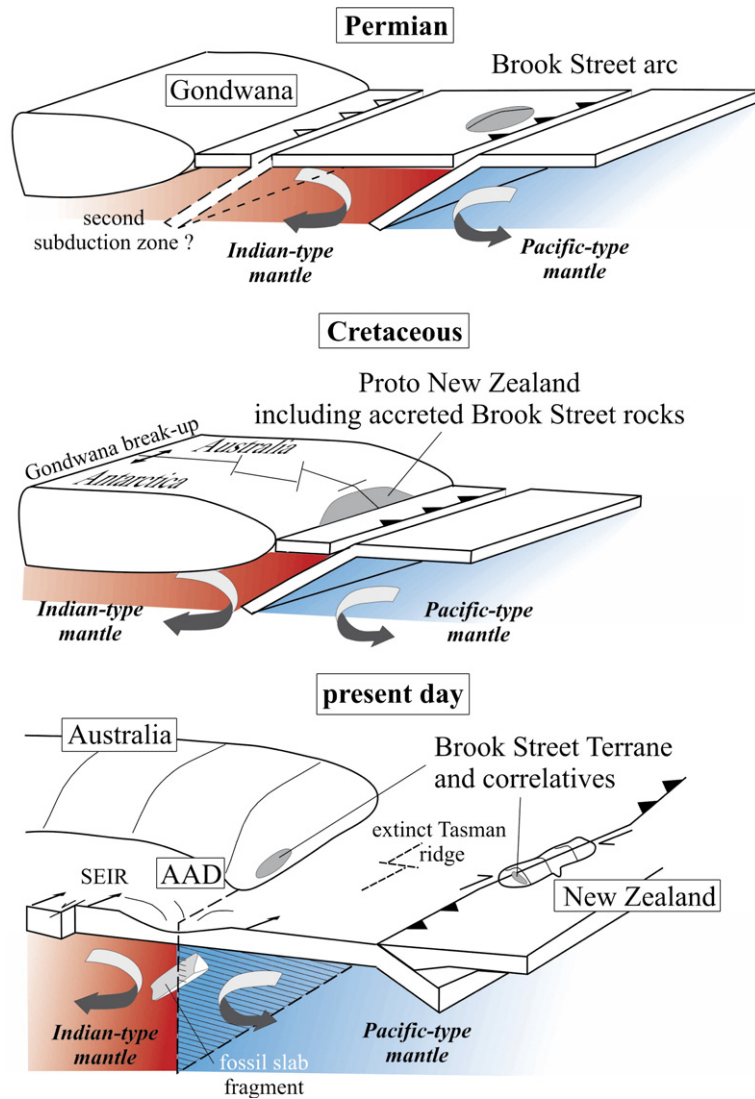


Fig. 9. Schematic sketches illustrating the paleo-tectonic development of the mantle domain boundary in the SW Pacific region since Permian time. SEIR=South East Indian Ridge.

existed since at least Permian time has far reaching implications for the understanding of timescales of convection-cell dynamics. Because both convecting reservoirs preserved distinct isotope patterns since at least Permian time, no detectable mass transfer across the boundary appears to have taken place since. Mixing of mantle domains across destructive plate boundaries at shallow upper mantle levels is hampered by the subducting slab that may act as a natural barrier. The long-term preservation of isotope signatures such as observed in this region supports geophysical mixing models promoting the idea that significant mass exchange between adjacent convecting regimes is limited [60,61]. Notably, continuous mass transfer by subduction pro-

cesses is not incorporated in such a model. A continuous addition of subduction components has significant impact on the Hf–Nd elemental and isotopic budget of the mantle wedge ([3], this study). Hence, mass transfer across convection cell boundaries by subduction zone processes appears to be a much more effective mechanism of mass exchange between neighboring convection cells than convective mass exchange at deeper mantle levels.

With respect to the origin of the Indian mantle domain, it seems likely that the contamination of the Indian mantle convection cell was caused by incorporating delaminated subcontinental lithospheric mantle from the greater Gondwana supercontinent (e.g., [19,62]). The isostatic immersion of the Gondwana

supercontinent and its underlying subcontinental lithospheric mantle into a convecting mantle could have caused the observed widespread contamination, also in the Tethyan region. Mixing material that was metasomatized by subduction components along destructive plate boundaries into the upper mantle appears to be a less likely mechanism. Such a model would require the flux of vast amounts of subduction components into the upper Indian mantle along the western Pacific rim. Taking the extent of the “Indian” mantle domain from the western Pacific rim (Fig. 1) to the South West Indian Ridge [9] and the age of Indian-type mantle, being at least of Carboniferous age in the central Tethyan region [25], the uniformity of the isotope anomaly requires an effective homogenization of the mantle domain.

## 5. Conclusions

By combining the trace element and isotope compositions of rocks from the Permian Brook Street arc on South Island, New Zealand, the boundary between the Pacific and Indian mantle domains can be identified beneath this subduction system in Permian time. This conclusion is based on contrasting, Pacific-type Pb isotope signatures and Indian-type Hf–Nd isotope compositions. The mantle domain signatures of the sub-arc mantle wedge and the downgoing slab can be used as tracers of subduction polarity, in this case leading to the conclusion that the Brook Street arc was facing towards the south-eastern Gondwana margin (Fig. 9). The isotopic discrimination of mantle domains proposed here for Permian arc rocks may also be applied to other late Palaeozoic suspect terranes in south-eastern Gondwana. Because the Brook Street Terrane is thought to be part of a large island arc system in the Palaeo-Pacific, different segments of this arc that were accreted to East Gondwana potentially have similarly contrasting isotope signatures.

Because the location of the Brook Street arc prior to late Cretaceous rifting at the eastern Gondwana margin is similar to that of the Cretaceous position of the AAD, we suggest that the late Palaeozoic to Mesozoic subduction systems represents the precursor of the AAD that is presently located at the South East Indian Ridge.

The distinct Palaeozoic isotope signatures imply that the two mantle domains already existed in the SW Pacific region 260 m.y. ago, which is in general agreement with Indian-type isotope signatures reported from Palaeozoic rocks from Chinese ophiolites [24,25]. In particular, the isotope signatures in the Brook Street rocks show that the isotopic mantle boundary has persisted from at least the Permian until today. As a consequence, it can be argued that associated mantle convection cells do not show any

evidence of significant isotopic exchange since the late Permian, excluding subduction processes in the uppermost boundary zone. This time interval corresponds nearly to the typical time of one convective mantle overturn period ( $\sim 250$  m.y., e.g., [63,64]). Consequently, if delaminated subcontinental lithosphere from the greater Gondwana continent has indeed been incorporated into the Indian convection regime [65], this material could have been efficiently homogenized with the upper Indian mantle. The delamination of continent-like material could thus be a plausible cause for the widespread isotope signatures that are typical for Indian-type mantle. An alternative scenario, where Indian upper mantle was contaminated with a pelagic sediment component by long-persisting subduction activity in the western Pacific region appears less likely. In either scenario, however, the Indian convection cell would have been isolated at least since the Palaeozoic, and preserved its pristine isotope character.

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## Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at [doi:10.1016/j.epsl.2006.11.046](https://doi.org/10.1016/j.epsl.2006.11.046).

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