Mechanism and timing of Pb transport from subducted oceanic crust and sediment to the mantle source of arc lavas

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1. Introduction

Magmatism at convergent plate margins is thought to occur in response to the release of aqueous fluid from the subducting plate into the overlying mantle wedge (e.g. Gill, 1981; Tatsumi, 1986). This fluid, derived from dehydration and breakdown of hydrous minerals and water incorporated in sediments and altered oceanic crust of the subducting plate, carries with it significant amounts of fluid-soluble elements, including Pb (e.g. Pearce and Peate, 1995; Hawkesworth et al., 1991). As a result, subduction zone lavas, and ultimately the continental crust, are enriched in these elements relative to the upper mantle (e.g. Miller et al., 1994; Chauvel et al., 1995). At higher temperatures and pressures, a hydrous melt phase may also be important in transferring incompatible and fluid-soluble elements from the subducting plate to the overlying mantle (e.g. Schmidt and Poli, 2003; Kessel et al., 2005; Plank et al., 2009). Material derived from the subducting oceanic crust and the overlying mantle in subduction zone magmas represent new additions from the mantle to the crust, whereas addition of material from subducting sediments, largely derived from the continental crust, represent recycling of continental material, some of which is returned to the deeper mantle. Element transfer at subduction zones is therefore an important process in the chemical differentiation of the Earth, and many previous studies have sought to determine the contribution of these various sources to the lavas erupted at continental plate margins (see reviews by McCulloch and Gamble, 1991; Hawkesworth et al., 1993; Davidson, 1996; Elliott, 2003).

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Pb isotopes are a sensitive tracer of sediment addition to the mantle, because most marine sediments have much higher Pb concentrations, and very different Pb isotope compositions to oceanic basalts and the upper mantle. High $^{207}\text{Pb}/^{204}\text{Pb}$ ratios in many arc lavas relative to mid-ocean ridge basalts are generally thought to result from addition of subducted sediment (e.g. Woodhead, 1989; Elliott et al., 1997; Ewart et al., 1998). Unfortunately, it is much more difficult to estimate the relative contribution from subducting oceanic crust and mantle wedge to the Pb budget of arc lavas, because in most arcs these two sources have very similar Pb isotope compositions. In contrast, in the northern Tonga Arc, a seamount chain is being subducted, and as a result the subducting oceanic crust has a very different Pb isotope composition to the upper mantle. We show that subduction lavas from this arc can give new insights into the relative contribution of subducted oceanic crust, sediment and upper mantle to the Pb budget of arc lavas, and also constrain the relative timing of transfer of Pb from subducting sediment and oceanic crust to their mantle source. This in turn gives information on the likely physical mechanisms by which sediment and oceanic crust contribute to arc magmatism.

2. Sample locations and analytical techniques

The Tonga–Kermadec Arc is located in the southwestern Pacific, north of New Zealand (Fig. 1). At the Tonga Trench, the Pacific Plate is being subducted westwards beneath the Indo-Australian Plate. The rate of convergence increases, and the thickness of the sediment layer entering the trench decreases from south to north. The subducting Pacific Plate carries with it an aseismic ridge, the Louisville Seamount Chain (LSC), the northwestern part of which has been subducted beneath the Tonga Arc. Previous geochemical studies of Tongan lavas (Regelous et al., 1997; Turner et al., 1997; Turner and Hawkesworth, 1997; Wendt et al., 1997; Ewart et al., 1998; Hergt and Woodhead, 2007) have shown that the Pb isotope signature of the LSC can be discerned in the lavas erupted on Tafahi and Niuatoputapu Islands at the northern end of the arc, and that this distinctive signature can be used to estimate the rate at which Pb is transported from its source in the subducting slab to the surface in arc lavas (approximately 2–3 Ma; Regelous et al., 1997; Turner et al., 1997; Wendt et al., 1997).

We carried out Sr, Nd and high-precision (double-spike) Pb isotope analysis of a suite of 19 lavas from the two northern Tonga islands of Tafahi and Niuatoputapu (Table 1). Major and trace element data for most of these same samples are given in Ewart et al. (1998), Regelous et al. (1997), Wendt et al. (1997) and Turner et al. (1997). The isotope analyses were carried out at Royal Holloway using whole-rock chips where available, and both chips and powders were leached in 6 M HCl for 1 h, then rinsed repeatedly with ultrapure water before digestion in HF–HNO$_3$. Pb was separated from the sample solution using 0.1 ml Sr Spec resin in HCl; the total procedural blank was below 50 pg, which is negligible given the amount of sample Pb processed (typically 200 ng). Pb isotope measurements were carried out using a $^{204}\text{Pb}$–$^{206}\text{Pb}$ double-spike, on an Isoprobe multicollector inductively coupled plasma mass spectrometer in dynamic mode, as described by Thirlwall (2002) and Thirlwall and Ankiewicz (2004). Repeat analyses of the NBS981 Pb isotope standard, treated as an unknown, gave $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ values of 16.9421±0.0028, 15.4996±0.0026, and 36.7214±0.0066 respectively ($n=10$, 2$r$) over the course of this study. Data in Table 1 have been normalised to the NBS981 values of Todt et al. (1996) for ease of comparison with existing data. Sr and Nd isotope measurements were carried out by thermal ionisation mass spectrometry on the same sample dissolutions using the “waste” from the Pb columns. Average values for the NBS987 Sr and Aldrich Nd isotope standards over the period of analysis were 0.710262±0.000009 and 0.511408±0.000004 (2$r$). The Sr isotope data in Table 1 have been normalised to a $^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.710250 for the NBS987 standard, and Nd data are reported relative to a value of 0.511856 for the La Jolla standard.

3. Results, and comparison with existing data

Lavas from Tafahi and Niuatoputapu have relatively uniform but distinct Sr and Nd isotope compositions. Tafahi lavas have $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of 0.703900 to 0.703981 and 0.512930 to 0.512959 respectively, whereas Niuatoputapu lavas have higher $^{87}\text{Sr}/^{86}\text{Sr}$ (0.703994 to 0.704097) and lower $^{143}\text{Nd}/^{144}\text{Nd}$ (0.512894 to 0.512904). The Sr and Nd isotope compositions of northern Tonga lavas lie at the enriched (high $^{87}\text{Sr}/^{86}\text{Sr}$, low $^{143}\text{Nd}/^{144}\text{Nd}$) end of the array defined by Tonga–Kermadec lavas. Sr, Nd and Pb have very different behaviour in melts and fluids in the subduction zone environment (e.g. Brenan et al., 1995; Keppler, 1996; Kessel et al., 2005). Since Sr and Nd data for Tafahi and Niuatoputapu lavas (including many of the samples analysed in this study) have been previously presented and discussed by Ewart et al. (1998), Turner et al. (1997) and Wendt et al. (1997), and since this paper is concerned with the origin of Pb in North Tonga lavas, we do not discuss the Sr and Nd isotope data in detail here. The new double-spike Pb data (Table 1) show that Tafahi and Niuatoputapu lavas also have distinct Pb isotope compositions, with lavas from the latter island having higher $^{206}\text{Pb}/^{204}\text{Pb}$ for a given $^{208}\text{Pb}/^{204}\text{Pb}$ (Fig. 2). Most of the samples analysed in this study have been analysed previously by conventional methods (Ewart et al., 1998; Hergt
and Woodhead, 2007; Regelous et al., 1997; Wendt et al., 1997; Pearce et al., 2007). In most cases, there is reasonable agreement between earlier data and the new double-spike data, although for Nd analyses marked with an asterisk, R1 indicates repeat analysis of same sample solution on different days with different instrument settings; R2 indicates repeat chemical processing and analysis of separate sample dissolution. Sr and Nd isotope measurements were made on a single sample dissolution, except for Nd analyses marked with an asterisk. 206Pb/204Pb for Nd double-spike for fractionation correction. Data are normalised to the NBS-981 values of Todt et al. (1996) for comparison with earlier studies. Sr, Nd and Pb isotope measurements were made on a single sample dissolution, except for Nd analyses marked with an asterisk. R1 indicates repeat analysis of same sample solution on different days with different instrument settings; R2 indicates repeat chemical processing and analysis of separate sample dissolution. Sr and Nd isotope measurements were carried out on a VG54s 5-collector mass spectrometer in dynamic mode, and data are normalised to values of 0.710250 and 0.511856 for the NBS-987 and La Jolla standards. Pb isotope measurements by MC-ICPMS in dynamic mode, using a mixed 204Pb–207Pb double-spike for fractionation correction. Data are normalised to the NBS-981 values of Todt et al. (1996) for comparison with earlier datasets. See text for more details.

4. Discussion

4.1. Subduction inputs of Pb to Tonga–Kermadec lavas

Previous studies have concluded that Pb isotope variations within lavas erupted along the Tonga–Kermadec Arc result from variable mixing of Pb derived from three main sources; subducted sediment, subducting oceanic crust, and the overlying mantle wedge (Ewart and Hawkesworth, 1987; Gamble et al., 1996; Regelous et al., 1997; Turner et al., 1997; Turner and Hawkesworth, 1997; Wendt et al., 1997; Ewart et al., 1998; Haase et al., 2002; Hergt and Woodhead, 2007; Pearce et al., 2007). In order to estimate the contributions of these sources to the Pb budget of Tonga–Kermadec lavas (Section 4.2), we require an accurate estimate of the isotope composition of Pb in each.
measured (present-day) Pb isotope compositions of these samples is considerably more radiogenic than young Pacific MORB, but less radiogenic than any of the northern Tonga lavas (Fig. 2).

The subducting Louisville Seamount Chain intersects the Tonga Trench at latitude 26°S (Fig. 1). The northwestern part of the LSC has been subducted beneath the Tonga Arc, and previous studies have concluded that arc lavas from the northern Tonga islands of Tafahi and Niuatoputapu have more radiogenic Pb isotope compositions which overlap with data for Louisville Seamount Chain lavas and volcaniclastics. Northerly material (tephras and the products of mass wasting), which is at least 54% of the Paci

The exact Pb isotope composition assumed for the mantle endmember will influence the proportions of Pb calculated for each component in northern Tonga lavas (Section 4.2.1.), but do not affect our conclusions regarding the timing and mechanisms of Pb transport discussed in Sections 4.4 and 4.5.

4.2. How much Pb is derived from subducting oceanic crust?

Previous studies (Regelous et al., 1997; Turner et al., 1997; Wendt et al., 1997; Ewart et al., 1998) have shown that the Pb isotope compositions of Tonga–Kermadec arc is much less well constrained. Previous studies have shown that both Pacific and Indian type mantle is present beneath different parts of the Lau Basin (Hergt and Hawkesworth, 1994; Peate et al., 2001; Hergt and Woodhead, 2007; Escrig et al., 2009). Also, most lavas from active spreading centres and from older crust in the Lau Basin have significantly lower Ce/Pb ratios than oceanic basalts, and have been contaminated by subduction-derived Pb. Lavas from the Central Lau Spreading Centre (Peate et al., 2001), which overlap with those of Indian MORB, but many of these lavas also have Ce/Pb<20, and therefore likely contain Pb derived from subducted sediment and/or oceanic crust. Additionally, more Pb isotope compositions of Pacific MORB and/or oceanic crust. The much less well constrained. Previous studies have shown that both Pacific and Indian type mantle is present beneath different parts of the Lau Basin (Chauvel et al., 1995; Miller et al., 1994). All data in Table 2 are measured values, and have not been age-corrected, since it is the present-day Pb isotope composition of the subducting material that is important in the calculations below.

Table 2

<table>
<thead>
<tr>
<th>Pb (ppm)</th>
<th>206Pb/204Pb</th>
<th>207Pb/204Pb</th>
<th>208Pb/204Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tonga sediment</td>
<td>19.42</td>
<td>15.60</td>
<td>39.28</td>
</tr>
<tr>
<td>Kermadec sediment</td>
<td>19.81</td>
<td>15.80</td>
<td>39.50</td>
</tr>
<tr>
<td>Subducting Pacific crust</td>
<td>19.50</td>
<td>15.50</td>
<td>39.00</td>
</tr>
<tr>
<td>Louisville basaltic crust</td>
<td>19.50</td>
<td>15.50</td>
<td>39.00</td>
</tr>
<tr>
<td>Louisville volcaniclastics</td>
<td>19.50</td>
<td>15.50</td>
<td>39.00</td>
</tr>
<tr>
<td>Pacific upper mantle</td>
<td>19.50</td>
<td>15.50</td>
<td>39.00</td>
</tr>
<tr>
<td>Lau Basin mantle</td>
<td>19.50</td>
<td>15.50</td>
<td>39.00</td>
</tr>
</tbody>
</table>

Weighted average Tonga sediment composition estimated from analyses of DSDP Site 204 and 595 sediment given in Ewart et al. (1998). Average Kermadec sediment composition calculated by combining data for DSDP Sites 595 and 275 from Ewart et al. (1998). Subducting Pacific oceanic crust composition is average of measured (present-day) values for unleached samples of Pacific crust close to the Tonga Trench from Worthington et al. (2006) and Castillo et al. (2009). Louisville basalt composition is the most radiogenic of the analyses of Louisville lavas in Cheng et al. (1987). Louisville volcaniclastic sediment composition is based on analyses of tuff/ash at DSDP Site 204 by Ewart et al. (1998). Pacific upper mantle is an average of data for fresh MORB from the northern East Pacific Rise from Niu et al. (1999), Regelous et al. (1999), Castillo et al. (2000), and Lau Basin mantle composition is average of analyses of Central Lau Spreading Centre lavas from Peate et al. (2001).
compositions of Tafahi and Niuatoputapu lavas extend towards the field defined by lavas and volcaniclastic sediments from the Louisville Seamount Chain (Fig. 2). Our new double-spike data show that Niuatoputapu lavas overlap with LSC basalt compositions, whereas the most radiogenic Tafahi lavas have lower 208Pb/204Pb for a given 206Pb/204Pb, compared to LSC lavas. The difference cannot be explained by a pelagic sediment component in these volcaniclastics, which would shift their compositions towards lower 206Pb/204Pb for a given 208Pb/204Pb, and higher 207Pb/204Pb, compared to Louisville lavas. It is possible that Tafahi lavas contain a small contribution from the "normal" Pacific oceanic crust underlying the LSC; in this case the isotope composition of the Pb derived from the subducting crust will lie on a LSC-PC tie-line in Fig. 4. Alternatively, it is possible that lavas from the older, subducted portion of the LSC have lower 208Pb/204Pb than those Louisville seamounts that have been sampled (only 8 of which have been analysed for Pb isotopes). The divergence of the Tafahi and Niuatoputapu data arrays at lower 206Pb/204Pb values suggests that the proportions of Pb derived from sediment and from the mantle are slightly different in each case.

The Tafahi and Niuatoputapu lavas with the most radiogenic Pb isotope compositions therefore contain Pb that is derived dominantly from the subducting oceanic crust. Since the subducting plate and upper mantle beneath the northern Tonga islands have very different Pb isotope compositions, mixtures of Pb derived from the mantle, the subducting oceanic crust, and subducted sediment will plot within the "mixing triangles" in Fig. 4, and the relative proportions of Pb from each source in each sample can be calculated from the position of the sample within these mixing triangles. These calculations show that sediment, Louisville oceanic crust and mantle wedge contribute approximately 0–26%, 42–90% and 8–37% respectively of the Pb in northern Tonga lavas (Table 3). The estimates obtained using the 208Pb/204Pb–206Pb/204Pb and 207Pb/204Pb–206Pb/204Pb systems are similar (Table 3). The amount of Pb derived from the subducting oceanic crust (which in North Tonga is the Louisville Seamount Chain) is therefore much greater than the amount of Pb derived from the mantle wedge. This conclusion is robust regardless of the exact mechanism of transfer of the Pb (whether by fluid or melt) from each source to the melting region. Note that if we had used an Indian MORB Pb isotope composition for the mantle wedge endmember, the calculated proportions of Pb derived from the LSC in Table 3 would be higher.

We recognise that the tectonic situation in northern Tonga is unusual, in that a seamount chain, rather than "normal" oceanic crust created at a spreading centre, is being subducted. An important consideration is therefore the extent to which the Pb flux inferred for northern Tonga is representative of other subduction zones. The northwestern end of the Louisville Seamount Chain is composed of relatively old (about 70 My), cold, and seawater-altered basaltic material, similar to the "normal" oceanic crust subducting at most subduction zones. Both are therefore likely to lose Pb by a similar
in the Pb isotope composition of the depleted mantle endmember. The lower 206Pb/204Pb endmember, which represents the upper mantle beneath northern Tonga. The latter cannot consist of uncontaminated de

The percentage of Pb derived from the oceanic crust (OCR, mantle) and subducted sediment (SED) in each sample was calculated from the position of the sample within the mixing triangle bounded by the three mixing endmembers, the Pb isotope compositions of which are given in Table 2. For Tafahi samples, an average volcaniclastic sediment was used as the OCR endmember, whereas for Niuatoputapu samples, the most radiogenic of the Louisville Seamount Chain lavas analysed by Cheng et al. (1987) was used. The two estimates for the contribution of each endmember are calculated using the 207Pb/204Pb of this assemblage increases with increasing phengite content, and the melt productivity of this assemblage becomes greater for the contribution of each endmember. However, the volcaniclastic sediments represent a relatively small reservoir of LSC Pb, compared to the Louisville Seamounts themselves, and a more likely explanation is that the radiogenic Pb in northern Tonga lavas is derived from the basaltic crust of the Louisville Seamount Chain itself, where the cover of pelagic sediment is lowest, and the thickness of Louisville-type crust is greatest. This interpretation is supported by the geographical distribution of lavas bearing the LSC signature, which is restricted to the islands of Tafahi and Niuatoputapu. It is not observed in lavas from the island of Fonualei to the south, nor in young submarine lavas north of Tafahi (with the possible exception of one sample from a submarine caldera about 50 km NW of Tafahi, Falloon et al., 2007).

4.3. Different processes transport Pb from subducted sediment and oceanic crust

The orientation of the data arrays in Fig. 4 allow us to place important constraints on the relative timing of Pb addition from different sources to the mantle source of northern Tonga lavas, and the processes by which this takes place. The slope of the linear arrays defined by Tafahi and Niuatoputapu lavas in Fig. 4 can be explained by mixing between a high 206Pb/204Pb LSC derived Pb component, and a lower 206Pb/204Pb endmember, which represents the upper mantle beneath northern Tonga. The latter cannot consist of uncontaminated Pacific or Indian upper mantle, because an extension of the Tafahi and Niuatoputapu data arrays to less radiogenic compositions intersects the South Tonga–Kermadec data array, but does not pass through the field defined by the majority of the Pacific or Indian MORB data (Fig. 3). The upper mantle must therefore contain a sediment contribution, which was added before the Pb derived from the LSC. Thus, sediment Pb must first have mixed with mantle Pb, and later Pb from the basaltic crust of the LSC was added separately. The new North Tonga data therefore show that Pb addition from subducting sediment and oceanic crust takes place at different times, and likely therefore by different processes, and also constrain the relative timing of Pb addition from these sources. The implications are discussed further below.

4.4. Mechanism of Pb transport from subducted materials

Turner et al. (1997) suggested that the LSC Pb signature in northern Tonga lavas results from melting of volcaniclastic sediments derived from the LSC. However, the LSC volcanics at Site 204 contain 1.2–1.4 ppm Pb (Ewart et al., 1998), and are overlain by 103 m of pelagic sediment containing an average of 30 ppm Pb. The high 206Pb/204Pb mixing endmember consists entirely of Louisville-derived Pb, with no contribution from pelagic sediment with lower 206Pb/204Pb, higher 207Pb/204Pb. This observation is difficult to explain by melting of mixed volcaniclastic–pelagic sediments, since there is no evidence for accretion of the younger (pelagic) sediments at the fore-arc. If volcaniclastic sediments release Pb at lower temperatures than pelagic sediment, it could be possible to selectively extract Pb from the former. However, the volcaniclastic sediments represent a relatively small reservoir of LSC Pb, compared to the Louisville Seamounts themselves, and a more likely explanation is that the radiogenic Pb in northern Tonga lavas is derived from the basaltic crust of the Louisville Seamount Chain itself, where the cover of pelagic sediment is lowest, and the thickness of Louisville-type crust is greatest. This interpretation is supported by the geographical distribution of lavas bearing the LSC signature, which is restricted to the islands of Tafahi and Niuatoputapu. It is not observed in lavas from the island of Fonualei to the south, nor in young submarine lavas north of Tafahi (with the possible exception of one sample from a submarine caldera about 50 km NW of Tafahi, Falloon et al., 2007).

The mixing relationships discussed in Section 4.3 indicate that beneath the northern Tonga Arc, Pb from sediment and from oceanic crust is added to the source of the arc lavas separately. Several recent trace element studies of arc lavas (Elliott et al., 1997; Class et al., 2000; Singer et al., 2007; Duggen et al., 2008), have also concluded that material from subducted oceanic crust and sediment are added to the source of arc lavas separately, and that the sediment component is likely added in the form of a melt. Sediment melting appears necessary to explain the recycling efficiency of non-fluid-mobile elements such as Th and Be (Nichols et al., 1994; Johnson and Plank, 1999). Schmidt and Poli (2003) suggested that aqueous fluids derived from dehydration of oceanic crust could potentially mobilise significant amounts of these elements (together with Pb) from the overlying sediments, but this process would result in addition of a fluid containing a mixture of Pb from sediment and oceanic crust to the overlying mantle, inconsistent with the mixing relationships observed in our data.

Phase relationships suggest that at depths of less than 100 km, sediments will undergo only limited fluid release, compared to the underlying basaltic oceanic crust (Schmidt and Poli, 2003). At greater (100–150 km) depths, both pelagic sediment and basaltic crust will be characterised by a similar mineral assemblage, consisting of garnet + clinopyroxene + phengite + coesite + kyanite ± rutile (e.g. Schmidt and Poli, 2003; Kessel et al., 2005). At temperatures above about 700 °C a solute-rich hydrous melt, rather than an aqueous fluid, will be in equilibrium with such an assemblage (Kessel et al., 2005; Hermann et al., 2006; Plank et al., 2008), and can more efficiently transport Pb into the overlying mantle wedge. The melt productivity of this assemblage increases with increasing phengite content, and
the degree of melting in the metasedimentary layer may therefore be approximately an order of magnitude higher in than in the underlying K-poor oceanic crust (Schmidt et al., 2004; Schmidt and Poli, 2003).

In summary, the existing experimental data indicate that subducting sediment and altered basalt are likely to release Pb-bearing aequous fluid or a hydrous melt at different depths within the subduction zone. Pb release from the basaltic portion of the slab is likely to take place during dehydration, and as discussed in Section 4.2, up to 90% of the Pb in North Tonga lavas is derived from the basaltic crust of the Louisville Seamount Chain. The main process that transfers mantle Pb to the arc crust beneath northern Tonga (and probably in other subduction zones over Earth history) is thus dehydration of the basaltic oceanic crust of the subducting plate.

4.5. Timing of Pb transport from subducted materials

As outlined above, the mixing relationships observed in North Tonga lavas apparently require Pb from subducted sediment and from the mantle to be mixed together before Pb from the oceanic crust is added, yet the available experimental data suggest that altered oceanic crust will lose Pb to the overlying mantle wedge by dehydration at lower pressures and temperatures than are required for significant release of Pb from sediment by melting. On the other hand, addition of fluid from dehydrating altered oceanic crust is most likely the process that ultimately triggers melting within the mantle wedge (e.g. Gill, 1981; Tatsumi, 1986), and is therefore expected to occur shortly before melting and magmatism.

A possible explanation for the observed mixing sequence is that the sediment signal is “old”, having been introduced into a convecting mantle wedge during previous subduction events. However, the presence of significant amounts of cosmogenic 10Be in Tonga–Kermadec lavas (including lavas from Tafahi; George et al., 2005), indicates that 10Be (and therefore also at least some Pb; Kessel et al., 2005) from pelagic sediment must have been introduced into the mantle source of these lavas relatively recently. The time taken for Be to be transported from trench to surface in lavas must be less than about 5 half-lives of 10Be, or 7.5 My. George et al. (2005) argued that the 10Be concentrations of Tonga lavas compared with the flux of 10Be into the Tonga Trench requires Be to have a “residence time” of about 4 My within the mantle wedge. If Be and Pb are released from sediment together (Kessel et al., 2005), then this therefore represents an upper limit on the time since Pb was added from the subducted basaltic crust of the LSC to the mantle source of north Tonga lavas.

In fact, the time taken for Pb from the subducting LSC to be transported through the subduction zone beneath northern Tonga can be directly estimated. Since the source of the Pb in northern Tonga lavas can be traced to a point on the subducting slab beneath these islands (i.e. the subducting LSC), then by making some assumptions regarding the depth at which Pb is likely to be released from the oceanic crust, Regelous et al. (1997) and Turner et al. (1997) showed that this observation can be used to calculate the absolute time taken for Pb to travel from its source in the subducting plate to the surface in lavas. For young Tafahi lavas, this transport time is estimated at 2–3 My (Regelous et al., 1997; Turner et al., 1997). However, most Niutatoputapu lavas are around 3 to 3.5 My old (Danyushhevsky et al., 1995), and so beneath this island, Pb from the slab appears to have taken considerably less time to reach the surface.

4.6. Comparison with U-series constraints

Additional constraints on the absolute timing of slab and sediment additions to the source of arc lavas come from short-lived U-series isotope data. A relatively late addition of at least part of the fluid component is required by uranium-series data for young Tonga–Kermadec lavas, most of which have \((^{238}\text{U}/^{230}\text{Th}) > 1\) (Regelous et al., 1997; Turner et al., 1997), which is also a characteristic of arc lavas generally (e.g. Elliott, 2003; Turner et al., 2003; Peate and Hawkesworth, 2005). Large \(^{226}\text{Ra}–^{230}\text{Th}\) disequilibrium in many arc lavas (including lavas from Tonga) indicates that some fluid addition occurred within \(< 10\) ky of eruption (Turner et al., 2001). However, the disequilibria in young arc lavas do not require that the entire budget of fluid-soluble elements was added to the mantle source within a few ky of melting and eruption. An indication of the time-integrated fluid addition can be obtained from Ba/Th and U/Nb ratios, because Ba and Nb have similar chemical behaviour to Ra and Pa respectively. For example, Ba/Th ratios of up to 900 (i.e. about 15 times greater than typical mid-ocean ridge basalts) together with \((^{226}\text{Ra}/^{230}\text{Th}) < 6\) in arc lavas worldwide (Turner et al., 2001) indicates that much of the Ba and Ra was added to the magma source > 5 half-lives of \(^{226}\text{Ra}\) ago. Similarly, Nb/U ratios of arc lavas are 10–20 times lower than typical upper mantle values, and require that 80–95% of the U in the former is fluid-derived (assuming that Nb/U is fractionated by fluid addition of U, rather than during melting).

A significant proportion of this U must be added to the wedge several half-lives of \(^{231}\text{Pa}\) (32,760 ky) before melting, in order for much of the excess \(^{235}\text{U}\) to decay so that melting generates magmas with \((^{231}\text{Pa}/^{235}\text{U}) > 1\) in the majority of subduction zones (Bourdon et al., 1999; Turner et al., 2006). U-series data for other arc lavas hint that sediment and slab-derived fluid are added to their source at different times; for example the Marianas lavas with the largest sediment component are closest to U–Th equilibrium (Elliott et al., 1997), suggesting that addition of the sediment component may have occurred several half-lives of \(^{230}\text{Th}\) ago.

4.7. Physical models for arc magmatism in North Tonga

The U-series constraints on fluid transport rates might be reconciled with the 2–3 My estimate for the time taken for Pb to travel from slab to surface if fluid-soluble elements such as Pb, U and Ra are transported from slab to mantle source by a series of hydration–dehydration reactions (Davies and Stevenson, 1992; Manning, 2004), over a period of time that is long compared to the half-lives of \(^{226}\text{Ra}\) and \(^{231}\text{Pa}\). In this model, Pb takes several My to be transported from the slab to the region of melting, and the latest dehydration event creates the U-series disequilibria, and triggers melting once the fluid reaches mantle that is hot enough to melt (Regelous et al., 1997). Alternatively, a flux melting model (Thomas et al., 2002), in which solid mantle traverses the melting region on timescales of 105–106 y, and is continuously fluxed by fluid from the slab, may better explain the presence of Louisville-derived Pb in lavas from northern Tonga erupted over a 3 My period.

The observation that sediment Pb is added to the mantle source recently (~4 My; George et al., 2005) and before Pb derived from the subducted oceanic crust by fluids, can be explained in at least two ways, as illustrated schematically in Fig. 5. One possible scenario (Fig. 5a) is that bulk sediment is physically mixed into the overlying mantle at relatively shallow depths, and this mixed source is then carried down to a depth at which the temperature of the oceanic crust is high enough that Pb-bearing fluid is released from the slab by dehydration, and triggers melting of the overlying mantle. Another possibility (Fig. 5b) is that sediments release Pb to the overlying mantle by melting at greater depths, followed by buoyant upwelling of this mixed source to shallower levels where fluids from the dehydrating oceanic crust induce melting (Elliott et al., 1997). Several recent studies have concluded that a sediment melt, rather than bulk sediment, contributes to arc magmatism (e.g. Elliott et al., 1997; Class et al., 2000; Singer et al., 2007). We therefore suggest that the scenario illustrated in Fig. 5b is the more likely.

4.8. Subducting crust Pb mass balance

Since we are able to estimate the proportion of Pb in northern Tonga lavas that is derived from subducting oceanic crust, we can
that the range in Pb flux calculated above for northern Tonga is representative of other convergent plate margins where "normal" oceanic crust (7 km thick, and containing 0.5 ppm Pb) is being subducted, then if Pb is extracted uniformly from the entire thickness of the crust, its Nd/Pb ratio will be decreased by approximately 3.5%, and a "subduction zone processing" signal would not easily be discerned in the Nd/Pb ratio of melts derived from this recycled crust if it were later to contribute to intraplate ocean island magmatism (Hofmann and White, 1982). A more likely situation is that Pb is removed from the upper parts of the oceanic crust, where thermal gradients are highest. In this case, more than half the thickness of the subducted oceanic crust could develop a resolvably (>5%) lower Nd/Pb ratio.

The proportion of Pb lost from the subducting oceanic crust estimated above represents a lower limit, since some Pb will be lost from the slab in the fore-arc and back-arc regions, where it does not contribute to arc magmatism.

5. Conclusions

In summary, the Pb isotope compositions of lavas from the northern Tonga islands of Tafahi and Niutaoputapu indicate that (a) the Pb budget of these lavas is dominated by the contribution from the subducting basaltic crust of the Louisville Seamount Chain, (b) transport of Pb by fluids from subducting oceanic crust is therefore the main mechanism by which Pb is transferred from mantle to crust at subduction zones, (c) mass balance calculations show that the amount of Pb transferred from oceanic crust to arc crust is equivalent to the Pb contained in a 67 to 143 m thick layer of the subducting Louisville crust (or up to 3.5% Pb removal from 7 km thick oceanic crust); these estimates place a lower bound on the amount of Pb lost from the subducting slab, (d) mixing relationships indicate that sediment Pb is mixed with mantle Pb, and then Pb from the subducting oceanic crust is added. The Pb from these two sources is therefore transported from the subducting plate into the overlying mantle by different processes.

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compare the mass of Pb entering the trench in the form of oceanic crust, to the mass of Pb transferred to the arc crust in the form of lavas and cumulates, in order to estimate the efficiency of mantle–crust transfer of Pb at subduction zones. An accurate mass balance is strictly possible only if a subduction zone is in "steady-state" (which is unlikely if a transient feature such as a seamount chain is being subducted), and also requires a knowledge of the amount of Pb that is lost from the subducting oceanic crust in the fore-arc and back-arc regions, which is unconstrained by our study. Nevertheless, our data can be used to place a lower bound on the amount of Pb that is lost from the subducting oceanic crust during subduction.

Northern Tonga lavas contain an average of 1.2 ppm Pb, of which 42–90% is derived from the subducting LSC (Table 3). Assuming that the primary melts of northern Tonga lavas contain 0.7 ppm Pb, and that the magma production rate at the Tonga Arc is 0.13 × 10⁶ kg/y/m (Bach et al., 1998), then the flux of Pb from the subducting LSC into the arc crust is 0.038 to 0.082 kg/y/m. Oceanic crust (density assumed to be 2800 kg/m³) is subducting at the northern end of the Tonga Trench at a rate of 0.24 m/y (Bevis et al., 1995), and contains approximately 0.85 ppm Pb (estimated from Nd concentrations of Louisville lavas, Cheng et al. (1987), assuming a Nd/Pb ratio of 25). The Pb flux from the subducting oceanic crust calculated above therefore corresponds to complete removal of Pb from an approximately 67 to 143 m thickness of the subducting Louisville crust.

Our mass balance calculation has implications for the composition of the oceanic crust that is subducted into the deeper mantle. Assuming

Fig. 5. Schematic diagrams showing two possible ways to explain the relative timing of Pb addition from subducting sediment and oceanic crust to the source of arc lavas, as inferred from Pb isotope mixing relationships in northern Tonga lavas. In (a), bulk sediment or sediment melt is mixed into the mantle at shallow levels (1), followed by upwelling of the hybrid mantle (2) to shallower depths, where addition of fluids from dehydration of the oceanic crust induces melting (3). Fig. 5b modified from Elliott et al. (1997).


