



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

Marcel Regelous^{a,b,*}, John A. Gamble^c, Simon P. Turner^d

^a Department of Earth Sciences, Royal Holloway University of London, Egham, Surrey TW20 0EX, U.K.

^b GeoZentrum Nordbayern, Universität Erlangen-Nürnberg, Schlossgarten 5, Erlangen 91054, Germany

^c Department of Geology, National University of Ireland, Cork, Ireland

^d GEMOC, Department of Earth and Planetary Sciences, Macquarie University, Sydney, NSW 2109, Australia

ARTICLE INFO

Article history:

Received 7 April 2009

Received in revised form 15 December 2009

Accepted 12 February 2010

Editor: B. Bourdon

Keywords:

Subduction

Tonga–Kermadec

Pb isotopes

Sediment

Mass balance

Oceanic crust

ABSTRACT

New high-precision (double-spike) Pb isotope analyses of lavas from Tafahi and Niuaotupapu, the northernmost islands of the Tonga–Kermadec Island Arc, are used to examine the source of Pb in these samples, and the relative timing of Pb addition from the subducting oceanic crust and subducting sediment. Lavas from these islands have distinctive, radiogenic Pb isotope compositions, which are inherited from the basaltic crust of the subducting Louisville Seamount Chain on the Pacific Plate. The subducting oceanic plate and the overlying upper mantle beneath northern Tonga therefore have very different Pb isotope compositions, allowing the proportion of Pb derived from each of these sources, and from subducting sediment to be estimated. We show that between 42% and 90% of the Pb in northern Tonga lavas is derived from the basaltic crust of the subducting Louisville Seamount Chain. The dominant source of mantle Pb in arc lavas (at least in northern Tonga), is therefore subducted basaltic oceanic crust, rather than the overlying mantle wedge. The orientation of the Pb isotope arrays constrain the relative timing of Pb addition from these different sources, and show that sediment Pb must be mixed with the upper mantle before Pb from the subducted oceanic crust is added during a separate event. This observation is consistent with the results of experimental studies, which suggest that altered oceanic crust and sediment are likely to lose Pb by dehydration or melting at different depths. Melting of sediment at depths >120 km, followed by migration of these melts to shallower levels within the overlying mantle, where Pb-bearing fluids derived from dehydration of oceanic crust trigger mantle melting, could explain the observed mixing relationships. Mass balance calculations show that the Pb flux into the arc magma source corresponds to the amount of Pb contained in the uppermost 67 to 143 m of the subducting basaltic crust of the Louisville Seamount Chain. If this minimum estimate is representative of subduction zones worldwide, where 7 km thick oceanic crust containing 0.5 ppm Pb is subducted, then the average Nd/Pb ratio of the oceanic crust that is recycled into the deep mantle is decreased by at least 3.5% as a result of subduction.

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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).

* Corresponding author. GeoZentrum Nordbayern, Universität Erlangen-Nürnberg, Schlossgarten 5, Erlangen 91054, Germany.

E-mail address: regelous@geol.uni-erlangen.de (M. Regelous).

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

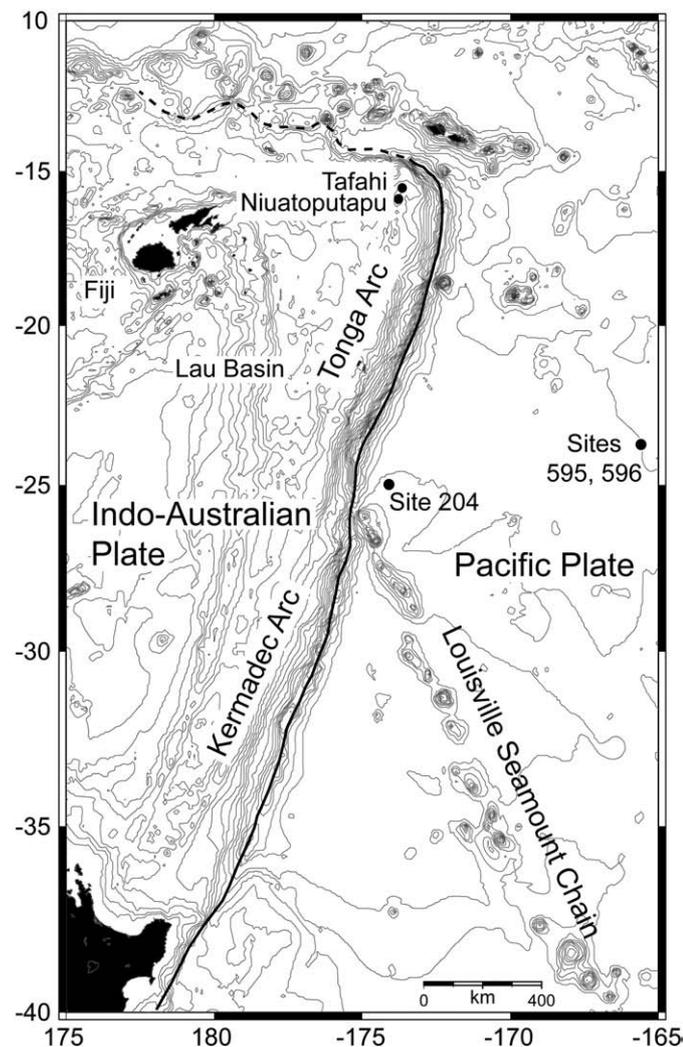


Fig. 1. Bathymetric map of the Tonga–Kermadec Arc, showing main tectonic features, and locations of samples analysed in this study. Also shown are the locations of DSDP Sites 204 and 595/596, and the position of the subducting Louisville Seamount Chain on the Pacific Plate.

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–HNO₃. Pb was separated from the sample solution using 0.1 ml Sr Spec resin in HCl; the total procedural blank was below 90 pg, which is negligible given the amount of sample Pb processed (typically 200 ng). Pb isotope measurements were carried out using a ^{204}Pb – ^{207}Pb double-spike, on an Isoprobe multicollector inductively coupled plasma mass spectrometer in dynamic mode, as described by Thirlwall (2002) and Thirlwall and Anckiewicz (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 ($n = 10, 2\sigma$) 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 (2σ). 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 samples 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 $^{208}\text{Pb}/^{204}\text{Pb}$ for a given $^{206}\text{Pb}/^{204}\text{Pb}$ (Fig. 2). Most of the samples analysed in this study have been analysed previously by conventional methods (Ewart et al., 1998; Hergt

Table 1
New Sr, Nd and double-spike Pb isotope data for lavas from the northern Tonga islands of Tafahi and Niuatoputapu.

Sample	Location	Type	$^{87}\text{Sr}/^{86}\text{Sr}$	$^{143}\text{Nd}/^{144}\text{Nd}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$
T068	Tafahi	P	0.703910 ± 10	0.512933 ± 4*	19.3278 ± 20	15.6149 ± 17	38.9469 ± 49
T068 R ₂	Tafahi	P	0.703924 ± 10				
T069	Tafahi	P	0.703900 ± 10	0.512957 ± 6			
T071	Tafahi	P	0.703904 ± 10	0.512949 ± 5*	19.0807 ± 34	15.6037 ± 29	38.7363 ± 78
T072	Tafahi	P	0.703923 ± 10		19.2722 ± 38	15.6142 ± 32	38.9046 ± 81
T072 R ₂	Tafahi	P	0.703916 ± 11				
T073	Tafahi	P	0.703951 ± 9	0.512959 ± 5			
T075	Tafahi	C	0.703914 ± 10	0.512930 ± 5	19.3442 ± 21	15.6171 ± 17	38.9592 ± 46
T075 R ₁	Tafahi	C	0.703917 ± 9				
T076	Tafahi	P	0.703903 ± 9	0.512957 ± 6	19.0324 ± 14	15.5874 ± 13	38.6749 ± 34
T113CP	Tafahi	C	0.703981 ± 10	0.512937 ± 5	19.3320 ± 26	15.6131 ± 21	38.9471 ± 55
T113CP R ₁	Tafahi	C	0.703980 ± 10		19.3332 ± 16	15.6143 ± 14	38.9508 ± 38
T116	Tafahi	C	0.703951 ± 9	0.512946 ± 4	18.9114 ± 17	15.5853 ± 15	38.5284 ± 41
T116 R ₁	Tafahi	C	0.703958 ± 9				
T051	Niuatoputapu	P	0.703994 ± 9	0.512894 ± 4	19.2454 ± 21	15.6082 ± 17	38.9330 ± 47
T052a	Niuatoputapu	P	0.704044 ± 9	0.512897 ± 4	19.0064 ± 24	15.5941 ± 21	38.7316 ± 63
T053	Niuatoputapu	P	0.704013 ± 10	0.512894 ± 5	19.2055 ± 24	15.6077 ± 20	38.9114 ± 54
T053 R ₂	Niuatoputapu	P			19.2070 ± 24	15.6092 ± 20	38.9146 ± 54
T054	Niuatoputapu	P	0.703998 ± 9	0.512895 ± 4*	19.2732 ± 19	15.6221 ± 16	38.9795 ± 44
T055	Niuatoputapu	P	0.704060 ± 12	0.512904 ± 4*	19.3012 ± 24	15.6171 ± 19	38.9899 ± 49
T057	Niuatoputapu	P	0.704015 ± 10	0.512896 ± 6*	19.0713 ± 21	15.5947 ± 18	38.7811 ± 49
T058	Niuatoputapu	P	0.704025 ± 9	0.512894 ± 5*			
T059	Niuatoputapu	P	0.704000 ± 10	0.512898 ± 4*	19.2732 ± 15	15.6220 ± 13	38.9794 ± 36
64-T-2	Niuatoputapu	C	0.704074 ± 9	0.512903 ± 4	18.9782 ± 24	15.5866 ± 20	38.6999 ± 53
64-T-2 R ₁	Niuatoputapu	C	0.704070 ± 10		18.9769 ± 19	15.5858 ± 16	38.6974 ± 43
64-T-8	Niuatoputapu	C	0.704097 ± 9	0.512902 ± 5	18.9237 ± 17	15.5861 ± 15	38.6485 ± 38
64-T-8 R ₁	Niuatoputapu	C			18.9236 ± 20	15.5859 ± 17	38.6481 ± 43

Samples were in the form of either 1–2 mm sized chips (C) or tungsten carbide milled powder (P); all samples were leached before dissolution. 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 carried out on a VG354 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 ^{204}Pb – ^{207}Pb 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.

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 the latter show less scatter, and there are some differences between our data and older data from the Open University (Turner et al., 1997), with the latter being systematically offset to lower $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$. Our samples (both chips and powders) were leached before analysis, and so sample alteration or contamination during crushing might explain some of the differences, however the consistent offset for both chips and powders suggests that analytical bias is more likely. We consider that our double-spike data are more accurate than previous data obtained using conventional TIMS methods. Three of the samples we have studied were also analysed by Hergt and Woodhead (2007) using the double-spike method. After normalising to a common value of the NBS981 standard, our data for T113 (Tafahi) and 64-T-2 (Niuatoputapu) are similar within error, but we obtain less radiogenic Pb isotope values for sample 64-T-8 than reported by Hergt and Woodhead (2007).

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.

The thickness and composition of sediments being subducted at the Tonga Trench are well-known from studies of sediments drilled at DSDP Sites 204, 595 and 596 (see Fig. 1; Menard et al., 1987; Zhou and Kyte, 1992; Turner et al., 1997; Ewart et al., 1998; Plank and Langmuir, 1998). At Site 596, approximately 70 m of pelagic and metalliferous clay, chert and hydrothermal sediment overlies Cretaceous basement. The topmost 103 m of sediment at Site 204 has similar characteristics, but is underlain by >44 m of tuffs and volcanoclastics derived from the Louisville Seamount Chain (Burns and Andrews, 1973). These volcanoclastics have over an order of magnitude lower Pb concentrations than the pelagic sediments (Turner et al., 1997; Ewart et al., 1998), and thus have little influence on the bulk Pb isotope composition of the sediment entering the Tonga Trench. The Pb isotope composition of the Site 595/596 bulk sediment given by Plank and Langmuir (1998) is based on a single analysis of surface sediment from a piston core by Ben Othman et al. (1989). However, Ewart et al. (1998) showed that the compositions of the youngest sediments at both Site 204 and Site 596 are influenced by tephra from the arc. The Ben Othman et al. (1989) average has a composition close to that of the youngest sediments at Site 204, suggesting that the relatively young sediment analysed in the latter study is indeed influenced by mantle-derived Pb from the arc, and are not representative of the bulk sediment pile. We have therefore estimated the weighted average Pb isotope composition of bulk Tonga sediment using the data given in Ewart et al. (1998) for samples from throughout the sediment column at both Site 204 and 596, and this new average (Table 2) is more radiogenic than the value given by Ben Othman et al. (1989) and Plank and Langmuir (1998).

The composition of the “normal” Cretaceous Pacific oceanic crust that is subducting beneath the northern Tonga Arc (Table 2) was calculated from data for unleached basaltic samples dredged from close to the trench by Worthington et al. (2006) and Castillo et al. (2009). It is possible that this sample set is biased towards younger, more alkalic lavas which were emplaced off-axis. The average of the

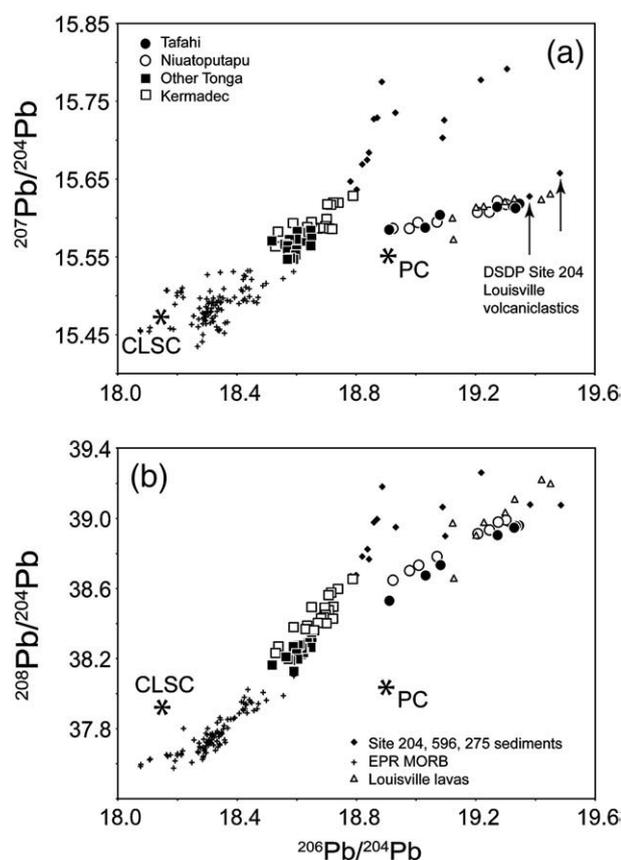


Fig. 2. Pb isotope compositions of Tonga–Kermadec lavas (data for Tafahi and Niuatoputapu lavas from this study, data for southern and central Tonga and Kermadec islands from Ewart et al., 1998). Data for Pacific sediments from DSDP Sites 204, 274, 596 (Ewart et al., 1998), Louisville Seamount Chain lavas (Cheng et al., 1987), and East Pacific Rise MORB (Regelous et al., 1999; Niu et al., 1999; Castillo et al., 2000) shown for comparison. Also shown are average compositions of Central Lau Spreading Centre lavas (CLSC: Peate et al., 2001) and subducting Pacific Cretaceous oceanic crust (PC: Worthington et al., 2006; Castillo et al., 2009). Most Tonga–Kermadec lavas have Pb isotope compositions that are intermediate between Pacific mantle and subducting sediment. Lavas from Tafahi and Niuatoputapu have more radiogenic Pb isotope compositions which overlap with data for Louisville Seamount Chain lavas and volcaniclastics. Note that mixtures of “normal” Pacific oceanic crust (PC), sediment, and either Pacific or Indian upper mantle are unable to explain the high $^{206}\text{Pb}/^{204}\text{Pb}$ ratios of northern Tonga lavas, and that a contribution from subducted Louisville basalts or volcaniclastics is therefore required.

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 contain Pb from subducted LSC material (Regelous et al., 1997; Turner et al., 1997; Turner and Hawkesworth, 1997; Wendt et al., 1997; Ewart et al., 1998). Basaltic lavas from the LSC have Pb isotope compositions that are much more radiogenic than those of Pacific MORB (Cheng et al., 1987). The LSC is surrounded by an apron of volcaniclastic material (tephras and the products of mass wasting), which is at least 44 m thick at DSDP Site 204. Two analyses of the Louisville-derived volcaniclastics from Site 204 by Ewart et al. (1998) have significantly lower $^{208}\text{Pb}/^{204}\text{Pb}$ for a given $^{206}\text{Pb}/^{204}\text{Pb}$ compared to the Louisville lavas analysed by Cheng et al. (1987). We have therefore calculated two Louisville endmember compositions (Table 2) based on the most radiogenic compositions obtained by Ewart et al. (1998) and Cheng et al. (1987), for Louisville volcaniclastic sediments and basaltic lavas respectively. These compositions lie at the radiogenic end of the range in

Table 2

Concentrations and isotope compositions of Pb of the endmembers used in mixing calculations.

	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	Pb (ppm)
Tonga sediment	18.82	15.67	38.87	45
Kermadec sediment	18.88	15.70	39.90	30
Subducting Pacific crust	18.91	15.55	38.16	0.3
Louisville basaltic crust	19.45	15.63	39.22	1.0
Louisville volcaniclastics	19.49	15.63	39.08	1.3
Pacific upper mantle	18.30	15.48	37.70	0.10
Lau Basin mantle	18.13	15.48	37.93	0.10

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).

Louisville compositions, and so the estimates of the proportion of Louisville-derived Pb in northern Tonga lavas in the following mixing calculations are likely to represent lower limits. We assume that hydrothermal processes may redistribute Pb but do not significantly change the Pb isotope composition of older oceanic crust (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.

The Pb isotope composition of the upper mantle beneath the northern 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 clearly been contaminated by subduction-derived Pb. Lavas from the Central Lau Spreading Centre (Peate et al., 2001), which is situated furthest from the active arc, have Pb isotope compositions (Table 2, Fig. 2) which overlap with those of Indian MORB, but many of these lavas also have $\text{Ce}/\text{Pb} < 20$, and therefore likely contain Pb derived from subducted sediment and/or oceanic crust. An additional complication is that southward flow of Samoa-type mantle may have modified the Pb isotope composition of the mantle beneath the northern Tonga Arc and North Lau Basin (Danyush-evsky et al., 1995; Wendt et al., 1997; Pearce et al., 2007; Regelous et al., 2008). The Pb isotope compositions of Lau Basin lavas are thus highly variable (e.g. Escrig et al., 2009), and in the following discussion we have used an average Pb isotope composition of northern East Pacific Rise lavas (Regelous et al., 1999; Niu et al., 1999; Castillo et al., 2000) to represent the composition of the upper mantle beneath northern Tonga. These datasets include both normal and enriched MORB types, from an area situated far from active intraplate magmatism. Inclusion of data for MORB from all Pacific spreading ridges expands the Pacific MORB field considerably (Fig. 3), because this dataset includes samples from “hotspot”-influenced ridge segments, but our estimate of the mantle Pb composition in Table 2 lies within the field defined by the bulk of the Pacific MORB data in Fig. 3. 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.), 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

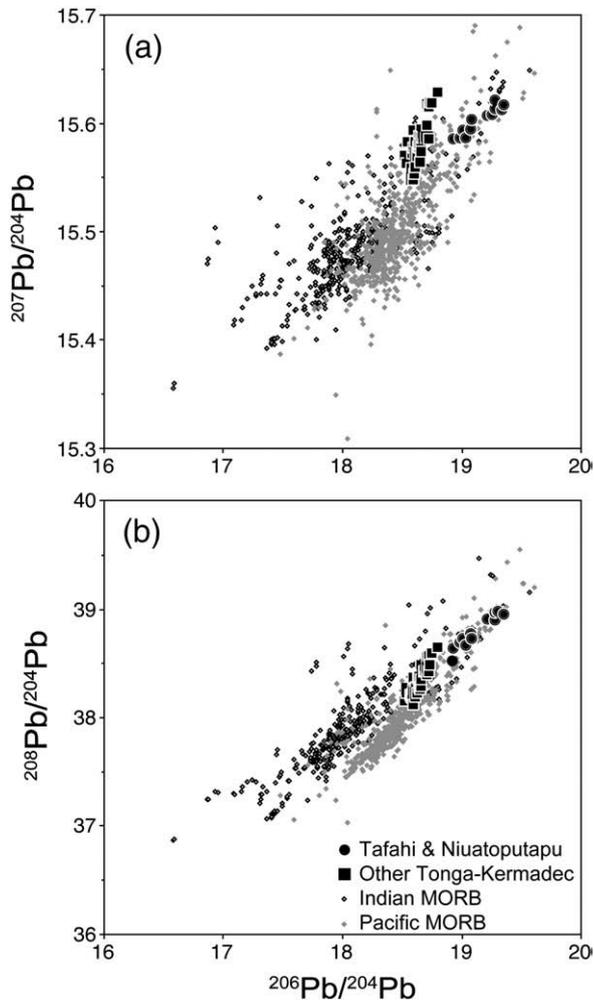


Fig. 3. Pb isotope compositions of Tonga–Kermadec lavas (data sources as above), together with MORB from Pacific and Indian Ocean spreading centres taken from the PETDB database. An extension of the North Tonga data array to less radiogenic compositions does not pass through the field defined by the majority of the MORB data in (a), indicating that these arrays do not result from binary mixing of Pb derived from the Louisville Seamount Chain and “normal” Pacific oceanic crust and mantle, but also require a sediment contribution.

compositions of Tafahi and Niuatoputapu lavas extend towards the field defined by lavas and volcanoclastic 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 $^{208}\text{Pb}/^{204}\text{Pb}$ for a given $^{206}\text{Pb}/^{204}\text{Pb}$, compared to LSC lavas. The difference cannot be explained by a pelagic sediment component in these volcanoclastics, which would shift their compositions towards lower $^{206}\text{Pb}/^{204}\text{Pb}$ for a given $^{208}\text{Pb}/^{204}\text{Pb}$, and higher $^{207}\text{Pb}/^{204}\text{Pb}$ for a given $^{206}\text{Pb}/^{204}\text{Pb}$, 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 $^{208}\text{Pb}/^{204}\text{Pb}$ 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 $^{206}\text{Pb}/^{204}\text{Pb}$ 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

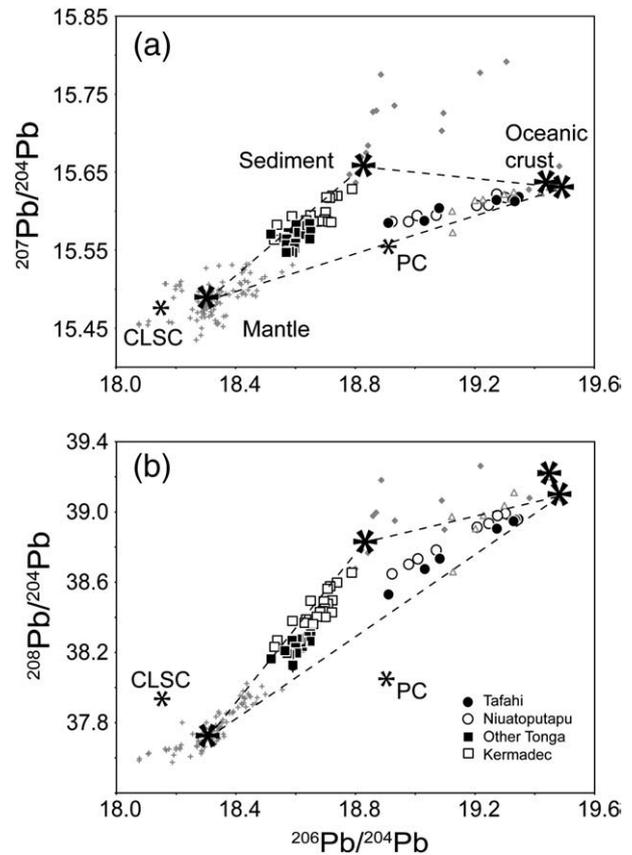


Fig. 4. Pb isotope compositions of Tonga–Kermadec lavas, and average endmember compositions used in modelling (bold star symbols). The contribution of each endmember to individual northern Tonga lavas can be calculated from the position of each sample within the mixing triangle (shown as dotted triangle for Tafahi samples), and the results are listed in Table 3. Two different Louisville endmember compositions were used, as described in the text. Also shown for reference (small star symbols) are average compositions of Central Lau Spreading Centre lavas (CLSC: Peate et al., 2001) and subducting Pacific Cretaceous oceanic crust (PC: Worthington et al., 2006; Castillo et al., 2009).

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 $^{208}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$ 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

Table 3
Estimates of sediment-, mantle-, and slab-derived Pb in northern Tonga lavas.

Sample	Location	OCR Pb (%) a	MAN Pb (%) a	SED Pb (%) a	OCR Pb (%) b	MAN Pb (%) b	SED Pb (%) b
T068	Tafahi	84.5	11.2	4.3	82.1	8.2	9.7
T071	Tafahi	56.7	23.0	20.3	55.5	21.4	23.1
T072	Tafahi	77.6	13.0	9.4	75.7	10.6	13.6
T075	Tafahi	85.8	9.8	4.4	84.0	7.5	8.5
T076	Tafahi	56.3	31.6	12.1	51.9	26.0	22.1
T113CP	Tafahi	85.7	11.9	2.4	82.8	8.3	8.9
T116	Tafahi	41.5	35.8	22.7	42.2	36.8	21.1
T051	Niuatoputapu	80.4	15.6	4.0	83.8	19.7	−3.4
T052a	Niuatoputapu	53.3	28.7	18.0	52.3	27.5	20.3
T053	Niuatoputapu	75.2	17.0	7.9	77.4	19.6	3.0
T054	Niuatoputapu	79.0	8.6	12.4	85.3	16.1	−1.4
T055	Niuatoputapu	84.7	10.0	5.3	90.2	16.7	−6.9
T057	Niuatoputapu	61.9	26.6	11.5	61.3	25.9	12.8
T059	Niuatoputapu	79.1	8.6	12.3	85.3	16.2	−1.4
64-T-2	Niuatoputapu	52.3	32.9	14.8	49.3	29.3	21.4
64-T-8	Niuatoputapu	45.1	34.7	20.3	42.6	31.7	25.7

The percentage of Pb derived from the oceanic crust (OCR), mantle (MAN) 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 volcanoclastic 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 $^{207}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$ (a) and $^{208}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$ (b) systematics, and are similar within 5% for most samples. The accuracy of these estimates may therefore be in the range of 5–10%, largely due to uncertainty in the Pb isotope composition of the depleted mantle endmember.

mechanism. We suggest that the greater thickness of the LSC crust compared to normal oceanic crust is unlikely to influence the proportion of Pb derived from the oceanic crust as inferred from Pb isotopes, because it is likely that Pb will be extracted from the uppermost part of the oceanic crust (where temperatures are highest during subduction), rather than from its entire thickness.

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 $^{206}\text{Pb}/^{204}\text{Pb}$ LSC derived Pb component, and a lower $^{206}\text{Pb}/^{204}\text{Pb}$ 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 been 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 volcanoclastic sediments derived from the LSC. However, the LSC volcanoclastics 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 $^{206}\text{Pb}/^{204}\text{Pb}$ mixing endmember consists entirely of Louisville-derived Pb, with no contribution from pelagic sediment with lower $^{206}\text{Pb}/^{204}\text{Pb}$, higher $^{207}\text{Pb}/^{204}\text{Pb}$. This observation is difficult to explain by melting of mixed volcanoclastic–pelagic sediments, since there is no evidence for accretion of the younger (pelagic) sediments at the fore-arc. If volcanoclastic sediments release Pb at lower temperatures than pelagic sediment, it could be possible to selectively extract Pb from the former. However, the volcanoclastic 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). This suggests that the Pb is derived from the basaltic crust of the LSC, which has a width of about 200 km, rather than the volcanoclastic apron each side of the LSC, which has a much greater extent. The Louisville seamounts that are being subducted at the Tonga Trench are 65–70 My old, and most thermal models of subduction zones do not predict significant melting of such old, cold basaltic crust (Peacock et al., 1994). Aqueous fluids are therefore likely responsible for transport of Pb from subducting basaltic crust or serpentinised peridotite. Aqueous fluid release from subducting altered basaltic crust will occur over a range of P , T conditions as continuous dehydration reactions consume hydrous phases, but major fluid release is likely to occur at the blueschist–eclogite facies transition at 15–25 kb (40–80 km), 500–600 °C (e.g. Schmidt and Poli, 1998; 2003; Hermann et al., 2006). In contrast, sediments may undergo only modest fluid release under these conditions due to the stability of phengite.

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., 2009), 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 aqueous 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 ^{10}Be in Tonga–Kermadec lavas (including lavas from Tafahi; George et al., 2005), indicates that ^{10}Be (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 ^{10}Be , or 7.5 My. George et al. (2005) argued that the ^{10}Be concentrations of Tonga lavas compared with the flux of ^{10}Be 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 Niutoputapu lavas are around 3 to 3.5 My old (Danyushevsky 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}Ra – ^{230}Th disequilibrium in many arc lavas (including lavas from Tonga) indicates that some fluid addition occurred within $\ll 10$ 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 basalt) 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}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}Pa (32,760 ky) before melting, in order for much of the excess ^{235}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}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}Ra and ^{231}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 10^5 – 10^6 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

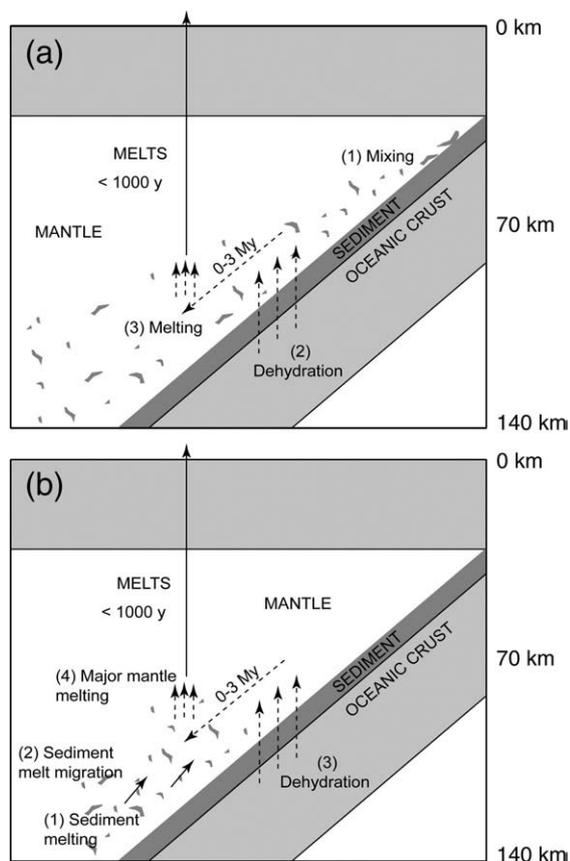


Fig. 5. Schematic diagrams showing two possible ways to explain the relative timing of Pb addition from subducting sediment and oceanic crust to the source of arc lavas, as inferred from Pb isotope mixing relationships in northern Tonga lavas. In (a), bulk sediment or sediment melt is mixed into the mantle at shallow levels (1), then carried to greater depth where fluids derived from dehydration of subducted oceanic crust (2) induce melting of the overlying sediment-contaminated mantle (3). In (b), melting of sediment takes place at greater depth (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).

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 0.13×10^6 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

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 oceanic 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 resolvable (>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.

Acknowledgements

We thank Cornelia Class and an anonymous reviewer for their constructive reviews, and Catherine Chauvel for comments on an earlier version of the manuscript. We thank Matthew Thirlwall and Christina Manning for help with the isotope measurements at Royal Holloway, and John Mahoney for sharing unpublished Pb isotope data for Louisville Seamount Chain lavas. S.P.T. acknowledges the support of an ARC Professional Fellowship. Pb isotope data for Pacific and Indian MORB in Fig. 3 were downloaded from the PetDB database.

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