University of Wollongong

Research Online

Faculty of Science, Medicine and Health -
Papers: part A

Faculty of Science, Medicine and Health

1-1-2014

Reappraisal of uranium-series isotope data in Kamchatka lavas: implications for continental arc magma genesis

Anthony Dosseto

University of Wollongong, tonyd@uow.edu.au

Simon Turner

Macquarie University

Follow this and additional works at: <https://ro.uow.edu.au/smhpapers>

Part of the [Medicine and Health Sciences Commons](#), and the [Social and Behavioral Sciences Commons](#)

Recommended Citation

Dosseto, Anthony and Turner, Simon, "Reappraisal of uranium-series isotope data in Kamchatka lavas: implications for continental arc magma genesis" (2014). *Faculty of Science, Medicine and Health - Papers: part A*. 1357.

<https://ro.uow.edu.au/smhpapers/1357>

Research Online is the open access institutional repository for the University of Wollongong. For further information contact the UOW Library: research-pubs@uow.edu.au

Reappraisal of uranium-series isotope data in Kamchatka lavas: implications for continental arc magma genesis

Abstract

Uranium-series isotopes can be used to determine constraints on the timescale of slab dehydration and melt production at subduction zones. However, interpretations of U–Th–Ra data suggest very different timescales of slab dehydration. Here, we present new U–Th–Ra data from Kamchatka along with a number of alternative models for production of radioactive disequilibrium. Variations in ($^{226}\text{Ra}/^{230}\text{Th}$) and ($^{231}\text{Pa}/^{235}\text{U}$) activity ratios are best explained by crystal fractionation with host rock assimilation for a duration of less than c. 6000 years. The association of the largest ^{226}Ra excesses with high Sr/Th in the most primitive lavas suggests that Ra–Th fractionation is controlled by slab dehydration less than 10 ka ago. We show that U–Th data can be explained by dynamic melting of a recently (<10 ka) metasomatized mantle wedge. Dynamic melting of an oxidized source metasomatized several hundreds of thousands of years ago cannot produce significant ^{231}Pa excess. Because ^{238}U – ^{230}Th disequilibrium is inferred to be controlled by partial melting, there is no requirement for multi-stage slab dehydration commencing ~150 ka. We suggest that Ra–Th disequilibria constrain the timing of slab dehydration, whilst U–Th fractionation is dominated by partial melting, at least at the Kamchatka arc.

Keywords

magma, arc, continental, implications, lavas, genesis, kamchatka, reappraisal, data, isotope, series, uranium

Disciplines

Medicine and Health Sciences | Social and Behavioral Sciences

Publication Details

Dosseto, A. & Turner, S. (2014). Reappraisal of uranium-series isotope data in Kamchatka lavas: implications for continental arc magma genesis. Geological Society Special Publication, 385 103-116.

Reappraisal of uranium-series isotope data in Kamchatka lavas: implications for continental arc magma genesis

Anthony Dosseto^{1,*} and Simon Turner²

¹ Wollongong Isotope Geochronology Lab, GeoQuEST Research Centre, School of Earth and Environmental Sciences. University of Wollongong. Northfields Avenue, Wollongong NSW 2522 Australia

² GEMOC Centre, Department of Earth and Planetary Sciences. Macquarie University. North Ryde NSW 2109 Australia

* corresponding author: tonyd@uow.edu.au

4120 words

62 references

2 tables

6 figures

Abstract

Uranium-series isotopes can be used to determine constraints on the timescale of slab dehydration and melt production at subduction zones. However, interpretations of U-Th-Ra data suggest very different timescales of slab dehydration. Here, we present new U-Th-Ra data from Kamchatka along with a number of alternative models for production of radioactive disequilibrium. Variations in ($^{226}\text{Ra}/^{230}\text{Th}$) and ($^{231}\text{Pa}/^{235}\text{U}$) activity ratios are best explained by crystal fractionation with host rock assimilation for a duration of less than ~6,000 years. The association of the largest ^{226}Ra excesses with high Sr/Th in the most primitive lavas suggests that Ra-Th fractionation is controlled by slab dehydration less than 10 ka ago. We show that U-Th data can be explained by dynamic melting of a recently (<10 ka) metasomatized mantle wedge. Dynamic melting of an oxidised source metasomatized several hundreds of thousands of years ago cannot produce significant ^{231}Pa excess. Because ^{238}U - ^{230}Th disequilibrium is inferred to be controlled by partial melting, there is no requirement for multi-stage slab dehydration commencing ~150 ka. We suggest that Ra-Th disequilibria constrain the timing of slab dehydration, whilst U-Th fractionation is dominated by partial melting, at least at the Kamchatka arc.

Over the past twenty years, uranium-series isotopes have been used to quantify the timescale of magmatic processes (e.g. Bourdon et al. 2003; Dosseto et al. 2010). This is because isotopes of the uranium decay chains fractionate during magma production and differentiation, and their abundance is time-dependent. Thus, radioactive disequilibrium in the ^{238}U - ^{230}Th - ^{226}Ra and ^{235}U - ^{231}Pa systems measured in volcanic rocks that have undergone little differentiation (or for which differentiation can be accounted for) can be modelled to constrain conditions of partial melting (e.g. melting rate, porosity; McKenzie 1985; Rubin and Macdougall 1988; Spiegelman and Elliott 1993; Sims et al. 1995; Sims et al. 1999; Asmerom et al. 2000; Spiegelman 2000; Zou and Zindler 2000; Landwehr et al. 2001; Sims et al. 2002; Bourdon et al. 2003; Dosseto et al. 2003; Rubin et al. 2005; Turner et al. 2005; Stracke et al. 2006; Petford et al. 2008; Sims et al. 2008; Bourdon and Elliott 2010). Radioactive disequilibrium in co-genetic volcanic rocks can also be used to model rates of crystallisation, since disequilibrium generally decreases over timescales similar to that of magma differentiation. Previous studies have inferred crystallisation rates ranging from 10^{-5} to $6 \times 10^{-4} \text{ yr}^{-1}$ (Sigmarsson 1996; Turner and Hawkesworth 1997; Turner et al. 1999; Cooper et al. 2001; Turner et al. 2001; Cooper and Reid 2003; Rogers et al. 2004; Blake and Rogers 2005; Peate 2005; Sigmarsson et al. 2005; Price et al. 2007; Touboul et al. 2007; Dosseto et al. 2008; Dosseto and Turner 2010). Moreover, in subduction zones, U-series isotope disequilibrium can be used to constrain the timing of slab dehydration under the volcanic arc (e.g. Elliott et al. 1997; Hawkesworth et al. 1997; Turner and Hawkesworth 1997; Turner et al. 1999; Turner et al. 2001; Thomas et al. 2002; Bourdon et al. 2003; Dosseto et al. 2003; Reagan et al. 2006; Tepley III et al. 2006; Huang et al. 2008; DuFrane et al. 2009): previous studies have argued that excess of uranium-238 (^{238}U) over thorium-230 (^{230}Th) in arc lavas requires that slab dehydration occurs over timescales of tens to hundreds of thousands of years (e.g. Turner and Hawkesworth 1997). However, the observation of radium-226 (^{226}Ra) excesses over thorium-230 (^{230}Th) suggests that slab-derived fluids have been added less than 10,000 years before eruption, implying fast melting rates and rapid magma transport towards the surface (10's of m/yr; Turner et al. 2001). In one model, the apparent discrepancy in ages of slab dehydration from ^{230}Th - ^{238}U and ^{226}Ra - ^{230}Th disequilibria was resolved by multi-stage slab dehydration, where ^{226}Ra - ^{230}Th disequilibrium reflect the last increments of fluid addition (Turner et al. 2000). In the Kamchatka arc, Dosseto et al. (2003) suggested that lava compositions are best explained by mixing between a melt derived from slab melting, that has a ^{230}Th excess, and a melt produced during partial melting of the metasomatized mantle, that has ^{238}U and ^{226}Ra excess. Based on the inferred end-member composition of the mantle-derived melt, they proposed that the mantle was hydrated during two episodes of slab dehydration at 150 ka and <10 ka. However, a subsequent study by Turner et al.

(2007) did not observe ^{230}Th excesses in the same Kamchatka lavas. Although the potential absence of ^{230}Th excess would not affect conclusions on the timing of slab dehydration and production of melts in the mantle, this could preclude the existence of a slab-derived melt and this has motivated us to re-visit the samples analysed by Dosseto et al. (2003). The new U-Th-Ra data are discussed from the perspective of the on-going debate on the apparent discrepancy between the slab dehydration timescales derived from ^{238}U - ^{230}Th and ^{226}Ra - ^{230}Th disequilibria (e.g. Turner et al. 2007; DuFrane et al. 2009; Avanzinelli et al. 2012).

Study area and geochemistry of samples

The Kamchatka Peninsula is located in the north-western corner of the Pacific Ocean, where the Pacific plate, with an age of 80-90 Ma, subducts under the Eurasian plate at rate of 9 cm/yr and with a dip angle of 49° (Fig. 1; DeMets et al. 1990). The Kamchatka arc is one of the most active volcanic arcs in the world, with the Kliuchevskoi volcano alone erupting $\sim 55 \times 10^6$ tons of magma per year (Melekestsev et al. 1991). Both tholeiitic and calc-alkaline lavas are produced, with lithologies varying from high-magnesian basalts to dacites and rhyolites. Radiogenic isotope compositions preclude any significant involvement of sediments in the source of the magmas (Kersting and Arculus 1995) but a significant contribution from a slab-derived fluid is inferred. This, along with the observation of enrichment in Large Ion Lithophile Elements (LILEs), suggests that the magmas are derived from partial melting of hydrated peridotite.

The samples analysed here are the same as those presented in Dosseto et al. (2003) and were collected within a 50-km diameter area from the Kliuchevskoi, Bezymianny and Tolbachik volcanoes (Fig. 1). These volcanoes are located in the Central Kamchatka Depression (CKD), one of the three volcanic regions of the peninsula. Samples analysed are two basalts from Kliuchevskoi, three from Tolbachik, one basic and two acid andesites from Bezymianny. Two of the basalts from Kliuchevskoi studied in Dosseto et al. (2003) were not available for analysis. The major, trace element and radiogenic isotope compositions were previously discussed in Dosseto et al. (2003). Sr and Nd isotope compositions suggest that studied rocks are derived from the same source (Dosseto et al. 2003). All samples are aphyric to subaphyric, except for sample PT30-4/1976 which is vesicular and has glomerocrysts of plagioclase and olivine (<1 cm). This sample also differs from the rest because it belongs to the high-K calc-alkaline series whereas other samples are calc-alkaline (following Miyashiro (1974) classification). It also has a different trace

element pattern shifted to higher incompatible element concentrations. Oxygen isotope data indicate the samples are devoid of any alteration by meteoric water (Pineau et al. 1999) .

Analytical techniques

Analyses were performed on the same powders as in Dosseto et al. (2003) (samples were not re-crushed). About 100 mg of powder was spiked with ^{236}U - ^{229}Th and ^{228}Ra tracers. Samples were digested in a mixture of HClO_4 , HCl , HNO_3 and HF . After driving off fluorides by evaporation at 100°C , residual HClO_4 was dried down by step heating at 150, 170 and 200°C . U, Th and Ra separation was performed using ionic chromatography techniques described in Dosseto et al. (2006) and Sims et al. (2008). U and Th concentrations and isotopic compositions were measured by multi-collector inductively-coupled plasma mass spectrometry (MC ICP-MS) on a Nu Instrument Nu Plasma at Macquarie University. Acquisition methods are described in Sims et al. (2008). MC ICP-MS yields beams of several volts for ^{232}Th and >1,000 counts per second (cps) for ^{230}Th , compared to only a few 10's of mV for ^{232}Th and <100 cps for ^{230}Th by TIMS using tungsten filaments in Dosseto et al. (2003)(Dosseto et al. 2003). Ra concentration was determined by thermal ionisation mass spectrometry (TIMS) on a Thermo Triton at Macquarie University. ^{226}Ra and ^{228}Ra were measured dynamically on a secondary electron multiplier. Accuracy and reproducibility of measurements were checked by analysis of the rock standard TML-3 and replicate analysis of two samples. For TML-3, $(^{234}\text{U}/^{238}\text{U}) = 1.003 \pm 0.003$ (2σ ; $n=10$), $(^{230}\text{Th}/^{238}\text{U}) = 0.994 \pm 0.006$ (2σ ; $n=10$), and the $^{230}\text{Th}/^{232}\text{Th}$ ratio deviates from the recommended value (5.816×10^{-6} ; (Sims et al. 2008)) by 0.5% ($n=10$). Replicate analysis of two samples is given in Table 1.

Results

When compared with the original data (Dosseto et al. 2003), significant discrepancies are evident (Table 1). Note that reported relative 2 standard errors are calculated by taking the worst replicate analysis for each parameter (for instance PT33-1/82 for $(^{226}\text{Ra}/^{230}\text{Th})$ but AXO3/84 for $(^{234}\text{U}/^{238}\text{U})$). The new results generally show a similar range of variation in U, Th and ^{226}Ra concentrations to those reported in Dosseto et al. (2003): between 0.333 and 1.035 ppm, 0.534 to 1.687 ppm and 188.6 to 676.8 fg/g, respectively. However, differences in U or Th concentrations up to 50% were observed for some samples. It is possible that in the initial study, the sample-spike homogenisation was not complete and/or samples were not thoroughly powdered. ^{231}Pa concentrations were not determined in this study,

however ($^{231}\text{Pa}/^{235}\text{U}$) activity ratios were re-calculated using the new U concentrations and the natural $^{238}\text{U}/^{235}\text{U}$ ratio of 137.88.

All samples are in secular equilibrium for ^{234}U - ^{238}U , confirming that they have not undergone significant low-temperature alteration, except for AXO3/84 which shows a slight depletion of ^{234}U relative to ^{238}U . ($^{226}\text{Ra}/^{230}\text{Th}$) activity ratios are also similar to those in Dosseto et al. (2003), with values between 0.98 and 3.17 (Fig. 2). Re-calculated ($^{231}\text{Pa}/^{235}\text{U}$) ratios based on new U concentrations but old ^{231}Pa abundances also show significant ^{231}Pa excess, as previously observed (Fig. 2). While we were not able to re-measure ^{231}Pa concentrations, the quality of the ^{231}Pa data is not a concern because the method employed to determine ^{231}Pa concentrations is similar to that used for ^{226}Ra concentrations, and old and new ^{226}Ra concentrations show a relatively good agreement. Like ^{226}Ra abundances, ^{231}Pa is determined by isotope dilution with ^{233}Pa , and although beam sizes at masses 263 ($^{231}\text{PaO}_2$) and 265 ($^{233}\text{PaO}_2$) were small (a few 100's cps), the measured 263/265 ratios were close to 1, ensuring good precision and accuracy. The most striking difference with the previous study is in the ($^{230}\text{Th}/^{238}\text{U}$) ratios, where the new data indicate only values less than 1 (i.e. ^{238}U excess). Although sample heterogeneity could explain part of these discrepancies, this cannot explain the shift from ^{230}Th to ^{238}U excess. This difference could be derived from the difficulties encountered by Dosseto et al. in measuring $^{230}\text{Th}/^{232}\text{Th}$ ratios by TIMS using tungsten filaments on a Finnigan MAT262: less than 100 cps of ^{230}Th and only a few mV of ^{232}Th were obtained during analysis, and satisfying precision and accuracy could be difficult to achieve considering the large $^{232}\text{Th}/^{230}\text{Th}$ ratio measured (compared to isotopic dilution, e.g. $^{228}\text{Ra}/^{226}\text{Ra}$ or $^{233}\text{PaO}_2/^{231}\text{PaO}_2$, where ratios are close to unity).

Discussion

Reappraisal of the origin of CKD magmas

As shown above, although Ra-Th data do not show major discrepancies between the new and old measurements, U-Th data are significantly different and in this section we re-visit the interpretations of (Dosseto et al. 2003) in the light of the new data.

^{230}Th excesses were previously observed and this was interpreted as indicating the involvement of a slab-derived melt. The new data show no evidence for ^{230}Th excess and hence there is no requirement for such a slab-derived melt. The range in observed U-Th-Ra-Pa compositions was previously explained by mixing between a slab-derived melt and a mantle-derived melt, based on correlations between daughter-parent activity ratios. Although the new measurements show no correlation between

($^{230}\text{Th}/^{238}\text{U}$) and other daughter-parent activity ratios, the positive correlation between ($^{226}\text{Ra}/^{230}\text{Th}$) and ($^{231}\text{Pa}/^{235}\text{U}$) is still observed (Fig. 2b; ($^{231}\text{Pa}/^{235}\text{U}$) ratios were re-calculated using new U concentrations, but samples were not re-analysed for ^{231}Pa). Accordingly, we suggest that these variations no longer reflect mixing between two primitive magmas with different origins (slab and mantle), but instead magma chamber processes whereby the most primitive magma is characterised by the largest radioactive disequilibria (see discussion below). Thus, the end-member composition of the mantle-derived melt in [Dosseto *et al.*, 2003], named *HMB* for “high-magnesia basalt” and characterized by the largest ^{226}Ra excess, represents the composition of the *primitive magma* (Fig. 2b) and still constrains the origin of CKD magmas. Similarly to the conclusions in Dosseto *et al.* (2003), it is apparent on Fig. 2b that flux melting cannot adequately explain the data as this will produce a decrease in ($^{231}\text{Pa}/^{235}\text{U}$) with increasing ^{226}Ra excess, whereas the inferred primitive magma is characterised by high ($^{226}\text{Ra}/^{230}\text{Th}$) and ($^{231}\text{Pa}/^{235}\text{U}$) ratios.

Rapid magma differentiation beneath the Central Kamchatka Depression

^{226}Ra - ^{230}Th and ^{231}Pa - ^{235}U disequilibria shows significant variations with Th concentrations (Fig. 3), used here as an index of magma differentiation. The broad negative correlations between the magnitude of these disequilibria and Th concentrations is interpreted as reflecting decay of radionuclides during differentiation along with fractionation and possible assimilation of host rock. This relationship can be used to place time constraints on the rate of magma differentiation in the crust. We use the model of assimilation and crystal fractionation presented in Dosseto *et al.* (2008). In this model, the change in activity ratios during differentiation is mostly controlled by the rate of crystallisation (in g/g/yr), and the assimilation/crystallisation ratio. Parameters used in the model are shown in Table 2. The maximum residual magma fraction allowed is 0.4 as below this value, magma viscosity may become too high to allow physical separation of newly formed crystals. The U, Th, Sr concentrations and ($^{226}\text{Ra}/^{230}\text{Th}$) and ($^{231}\text{Pa}/^{235}\text{U}$) activity ratios in the parental magma were chosen to best reproduce observed compositions. Note that the model is relatively insensitive to the values taken for U, Th, Pa and Ra bulk partition coefficients (assuming realistic values), even in the case of crystallisation of mineral phases such as amphibole, which is known to preferentially incorporate Ra over Th. In the case where no assimilation occurs, it is not possible to produce Th concentrations higher than 1ppm. Hence, we consider the scenario where the host rock is partially melted and assimilated into the differentiating magma. We assume that the assimilant (crustal melt) has the following composition: Th,

193 U and Sr concentrations are 100, 60 and 100 ppm, respectively, and $(^{226}\text{Ra}/^{230}\text{Th}) = (^{231}\text{Pa}/^{235}\text{U}) = 1$. High
 194 U and Th concentrations in the assimilant are required such that both Sr/Th and $(^{231}\text{Pa}/^{235}\text{U})$ data can be
 195 reproduced simultaneously. In order to keep the residual magma fraction at realistic values (≥ 0.4), a
 196 minimum assimilation/crystallisation ratio, r , of 0.1 is required to produce observed Th concentrations.
 197 The maximum r value considered is 0.3 (Tegner et al. 2005). Modelled curves calculated with r values
 198 between 0.1 and 0.3 yield similar fits to the data. Because the model is under-constrained, only a
 199 minimum crystallisation rate of 5×10^{-6} g/g/yr can be determined. If the crystallisation rate was lower
 200 than this value, $(^{226}\text{Ra}/^{230}\text{Th})$ ratios would decrease much faster than observed. This is used to infer a
 201 maximum differentiation timescale of 6,000 years to evolve from the most primitive to the most evolved
 202 compositions observed. Figure 3 shows an example of differentiation model curves where the
 203 crystallisation rate is set to 5×10^{-6} g/g/yr and $r = 0$ or 0.3. In this case, the differentiation timescale is less
 204 than 6,000 years (PT30-4/1976 is ruled out of the modelling as it is shown above that this sample is an
 205 outlier and is probably not genetically related to the other samples). Note that andesites deviate from
 206 the relationship formed by basalts in $(^{231}\text{Pa}/^{235}\text{U})$ versus Th concentration, while both andesites and
 207 basalts form a unique correlation in other diagrams. This deviation is not well understood and could
 208 indicate that andesites and basalts are not genetically related, where the source of the former would be
 209 characterised by a higher $(^{231}\text{Pa}/^{235}\text{U})$ (possibly reflecting a lower melting rate).
 210 It is shown here that crustal assimilation is required to account for the geochemical characteristics of
 211 Kamchatka lavas. Crustal assimilation has not been previously considered because of the lack of
 212 variation between Sr isotopes and indices of differentiation. However, because the crust assimilated is
 213 young its Sr isotopic composition may be similar to that of the newly emplaced magmas and the lack of
 214 Sr isotope variation does not justify ruling out crustal assimilation.
 215 Note that the ^{230}Th - ^{238}U disequilibrium does not show any systematic variations with indices of
 216 differentiation. A correlation is expected since this radioactive system operates on the same timescales
 217 as the ^{231}Pa - ^{235}U system and is equally sensitive to crystal fractionation and assimilation. This could be
 218 the result of (i) smaller variations in $(^{230}\text{Th}/^{238}\text{U})$ compared to $(^{226}\text{Ra}/^{230}\text{Th})$ and $(^{231}\text{Pa}/^{235}\text{U})$, such that the
 219 effect of assimilation is less clear than for the other radioactive systems; and (ii) conditions of partial
 220 melting of the host rock are such that the $(^{226}\text{Ra}/^{230}\text{Th})$ and $(^{231}\text{Pa}/^{235}\text{U})$ ratios of the crustal melt are
 221 significantly different than those of the primary magma (i.e. in secular equilibrium), whilst this would not
 222 be the case for $(^{230}\text{Th}/^{238}\text{U})$ ratios. This could be possible for instance if the porosity during partial
 223 melting of the crust was significant (e.g. $>1\%$) such that no or little ^{226}Ra - ^{230}Th and ^{231}Pa - ^{235}U disequilibria
 224 could be produced, while generating ^{230}Th - ^{238}U disequilibrium.

225

226 **Fluid origin of the ^{226}Ra - ^{230}Th disequilibrium**

227

228 High Ba/Th ratios are commonly used as an indicator of slab fluid addition. However a positive
 229 correlation between ($^{226}\text{Ra}/^{230}\text{Th}$) and Ba/Th does not unequivocally require a fluid origin for ^{226}Ra
 230 excesses because Ba is more incompatible than Th and, under certain conditions, decreasing degrees of
 231 partial melting could also lead to increases in Ba/Th ratios. Contrastingly, because Sr is less compatible
 232 but more fluid mobile than Th, an increase in Sr/Th ratios can only be explained by fluid addition and a
 233 positive correlation with ($^{226}\text{Ra}/^{230}\text{Th}$) would imply that (i) ^{226}Ra excesses are fluid-derived and (ii) less
 234 than 10 ka elapsed between slab dehydration and eruption. Such a correlation was observed by
 235 (Dosseto et al. 2003) and used to argue for a fluid origin of the ^{226}Ra - ^{230}Th disequilibrium. Our new
 236 measurements confirm the positive correlation (Fig. 3b). However, as shown in the previous section, the
 237 range of ($^{226}\text{Ra}/^{230}\text{Th}$) and Sr/Th values is explained by magma differentiation. It is important to note that
 238 whilst the positive correlation in Fig. 3b is accounted for by differentiation (cf. Huang et al. 2008),
 239 metasomatisation of the magma source is still required to produce the high ($^{226}\text{Ra}/^{230}\text{Th}$) and Sr/Th
 240 ratios in the most *primitive* magmas. Huang et al. (2011) invoked partial melting to account for ^{226}Ra -
 241 ^{230}Th disequilibrium in Kick'Em Jenny lavas (Lesser Antilles). However, if high ($^{226}\text{Ra}/^{230}\text{Th}$) ratios were
 242 produced during partial melting, they should be associated with low Sr/Th ratios (because the
 243 incompatibility order of these different elements is $\text{Ra} \gg \text{Th} > \text{Sr}$). Thus, the primary ^{226}Ra - ^{230}Th
 244 disequilibrium at the Kamchatka arc, and in arc lavas in general, is most likely derived from magma
 245 source metasomatism due to slab dehydration. The preservation of this ^{226}Ra - ^{230}Th disequilibrium
 246 implies rapid magma generation and fast transport to the surface (Turner et al. 2001).

 247 **Magma genesis at subduction zones: an alternative hypothesis to multi-stage**
 248 **fluid addition**

249 It is widely accepted that ($^{238}\text{U}/^{230}\text{Th}$) > 1 in arc lavas reflects metasomatism of the mantle wedge by
 250 slab-derived fluids (e.g. Turner et al. 2003). This contrasts with ($^{238}\text{U}/^{230}\text{Th}$) ratios < 1 produced during
 251 decompression melting (e.g. MORB and OIB) due to the greater incompatibility of Th relative to U. The
 252 timescales of fluid addition derived from ($^{238}\text{U}/^{230}\text{Th}$) > 1 are typically of the order of several tens of
 253 thousands of years (150 ka in the case of Kamchatka; Turner et al. 1998). However, ^{226}Ra - ^{230}Th
 254 disequilibrium is also observed in most arc lavas. Because any ^{226}Ra - ^{230}Th disequilibrium would decay

after 10 ka, and this disequilibrium is attributed to slab dehydration, the combined observations from ^{226}Ra - ^{230}Th and ^{238}U - ^{230}Th systems have been interpreted in some models to reflect different stages of slab dehydration. Although it is anticipated that there might be more than one episode of slab dehydration (because of the numerous hydrous phases being subducted) it has proved harder to identify which hydrous phases are linked to specific disequilibria.

Several models have been developed to explain how magmas are produced in the mantle wedge (see Bourdon et al. 2003 for a review). For instance, Thomas et al. (2002) have proposed a *flux melting* model where melts are continuously produced while slab-derived fluids are added to the mantle wedge. However, as shown above and in Dosseto et al. (2003), this model cannot account for observed ^{226}Ra - ^{230}Th and ^{231}Pa - ^{235}U disequilibria (Fig. 2b). In this section, we investigate whether dynamic melting following recent fluid addition to the mantle wedge (<10 ka ago) could account for the observed ^{238}U - ^{230}Th isotope compositions in Kamchatka. Dynamic melting involves a non-static source where matrix upwelling occurs (McKenzie 1985). In the case of subduction zones, we can consider that the source is advecting either as a result of fluid addition to the mantle wedge, creating a source that is more buoyant than the surrounding dry mantle, or as a result of downward convection along the slab (Dosseto et al. 2003). In our model, slab dehydration supplies U to magma sources shortly before melting (<10 ka ago), yielding a range in U/Th ratios at a constant ($^{230}\text{Th}/^{232}\text{Th}$). Assuming an upwelling or downwelling rate of 3 cm/yr, a porosity of 1‰ and a final degree of partial melting of 20% (see Fig. 4 caption for other parameters), it is possible to reproduce the positive array observed on the ($^{230}\text{Th}/^{232}\text{Th}$) vs. ($^{238}\text{U}/^{232}\text{Th}$) diagram (Fig. 4a). In this model, there is no demand for a multi-stage fluid addition to account for the different timescales of fluid addition inferred from ^{238}U - ^{230}Th and ^{226}Ra - ^{230}Th . Melts produced have ($^{231}\text{Pa}/^{235}\text{U}$) activity ratio between 1.49 and 1.58 (Fig. 4b), which is comparable to (but somewhat lower than) the composition of the most primitive sample. To reproduce the highest ($^{231}\text{Pa}/^{235}\text{U}$) observed, one could consider that the source starts with a higher ($^{231}\text{Pa}/^{235}\text{U}$) as a result of the presence of Pa in the fluid (Avanzinelli et al. 2012). However, this would require the source to have ($^{231}\text{Pa}/^{235}\text{U}$) ratios between 1.24 and 1.44. This would suggest that more Pa than U is carried in the fluid, which is unrealistic (e.g. Bourdon et al. 2003). Assuming that the metasomatized source has a ($^{226}\text{Ra}/^{230}\text{Th}$) of 3 (similar to the highest value observed in our samples), melting has little effect on this activity ratio, resulting in a slight increase to value of 3.2.

The model proposed above can explain U-series isotope compositions at other continental arcs. For instance, U-Th data of low-Ti lavas from Nicaragua (Reagan et al. 1994) can be reproduced using an up/downwelling rate of 6-7 cm/yr, a range of ($^{238}\text{U}/^{232}\text{Th}$) values for the source prior to melting between

1.8 and 2.7, a final degree of partial melting of 8% and a source mineralogy composed of 52% olivine, 20% orthopyroxene, 20% clinopyroxene and 8% garnet (Fig. 5a). This model also yields ($^{231}\text{Pa}/^{235}\text{U}$) values between 1.48 and 1.70, which is comparable to the range measured by Thomas et al. (2002) for Nicaraguan lavas (1.29-1.74). Similarly, U-series data from the Parinacota volcano (Bourdon et al. 2000) can be re-interpreted with this model: in Fig. 5b, ($^{230}\text{Th}/^{232}\text{Th}$) and ($^{238}\text{U}/^{232}\text{Th}$) ratios can be explained by dynamic melting of a recently metasomatized source to which sediments have been previously added in order to lower the source ($^{230}\text{Th}/^{232}\text{Th}$) ratio to 0.3. Melting parameters are similar to those used for the Nicaragua data, except for the up/downwelling rate (5 cm/yr). Note that in all cases the up/downwelling rates inferred are lower than plate convergence rates. If these up/downwelling rates reflect convection in the mantle wedge, this implies an incomplete coupling between the subducting slab and the overlying mantle.

It has recently been proposed that high-Mg basalts and andesites can be produced by partial melting of subarc peridotite and pyroxenite veins (Straub et al. 2011). Thus, an alternative model would be to consider a pyroxenite source. In this scenario, considering a source with 15% olivine, 80% orthopyroxene and 5% clinopyroxene (England and Davies 1973), it is possible to reproduce the ($^{238}\text{U}/^{232}\text{Th}$)-($^{230}\text{Th}/^{232}\text{Th}$) composition of Kamchatka rocks by simply considering a faster up/downwelling rate (4.5 cm/yr, which is still realistic considering the convergence rate is 9 cm/yr).

Another alternative model has been recently proposed where partial melting occurs under high oxygen fugacity (Beier et al. 2010). In this case, uranium becomes more incompatible than thorium (Lundstrom et al. 1994). Thus, the U-series isotope composition of lavas from the Manus basin were explained by dynamic melting in the back-arc of a mantle previously metasomatized and oxidised by slab-derived fluids. In principle, this model can be used to reproduce the U-Th isotope composition of Kamchatka lavas. Because fluid addition occurs >350ka ago, the metasomatized source has a high and variable ($^{230}\text{Th}/^{232}\text{Th}$) prior to melting (Fig. 6a). However, although significant ^{226}Ra - ^{230}Th disequilibrium can be produced in this model ($(^{226}\text{Ra}/^{230}\text{Th}) \sim 3$), no ^{231}Pa - ^{235}U disequilibrium is produced (because $D_{\text{U}}/D_{\text{Pa}}$ is only ~ 4 , versus ~ 100 in other models) (Fig. 6b). This is at odds with the observation of significant ^{231}Pa excesses in the most primitive Kamchatka lavas (note that there are at present no ^{231}Pa data with which to test this model for the Manus Basin).

Conclusions

In the light of recent results by Turner et al. (2007), the U-Th-Ra isotope composition of volcanic rocks from the Central Kamchatka Depression was re-visited. We conclude that:

- (i) New measurements show no ^{230}Th excess relative to ^{238}U , suggesting that there is no longer any evidence for a slab-derived melt.
- (ii) Variations in U-Pa and Th-Ra isotope compositions are best explained by magma differentiation with crustal assimilation, rather than mixing between slab- and mantle-derived melts as initially suggested (Dosseto et al. 2003). A maximum duration of magma differentiation of $\sim 6,000$ years can be inferred, with a minimum crystallization rate of 5×10^{-6} g/g/yr. Although not suggested by long-lived radiogenic isotopes (because the assimilant may have the same isotopic composition as the magma differentiating), assimilation is required and occurs at a minimum rate of 0.5×10^{-6} g/g/yr.
- (iii) High ($^{226}\text{Ra}/^{230}\text{Th}$) and Sr/Th ratios in the most primitive lavas emphasize that ^{226}Ra - ^{230}Th disequilibrium in arc lavas is derived from dehydration of the subducted slab.
- (iv) Multiple fluid additions to the mantle wedge are no longer required to account for U-Th-Pa-Ra isotope data. Ra-Th suggests recent fluid addition (< 10 ka ago) whereas U-Th data can be reproduced by dynamic melting of a metasomatized source, enriched in U over Th prior to melting. This is in agreement with the conclusions of Dosseto et al. (2003) who proposed that the primitive melt is produced by equilibrium porous flow or dynamic melting of a recently metasomatized mantle. Thus, U-Th-Pa-Ra data at the Kamchatka arc can be explained by a simple model where a single episode of fluid addition occurs shortly ($< \text{few thousand years}$) before dynamic melting of the upwelling or downwelling source. This model can also account for U-series isotope data at other continental arcs (Andes, Nicaragua). In addition, the observation of high ($^{226}\text{Ra}/^{230}\text{Th}$) associated with high ($^{231}\text{Pa}/^{235}\text{U}$) ratios confirm that flux melting is not adequate to model magma genesis beneath the Kamchatka arc.
- (v) An alternative model of arc magma production where the source is metasomatized more than 300,000 years ago and melting occurs under high oxygen fugacity cannot account for the observed ^{231}Pa - ^{235}U disequilibrium in primitive magmas.

Acknowledgements

We would like to thank the editors for putting this Special Publication together, Georg Zellmer for editorial handling and Riccardo Avanzinelli and Ian Smith for their insightful reviews. We would also like to thank Tracy Rushmer, Hugh O'Neill, Gerhard Wörner, Bernard Bourdon and Bruce Schaefer for helpful

350 discussions. The work was funded by an Australian Research Council Discovery Grant DP0988658.

351 Dosseto acknowledges an Australian Research Council Future Fellowship FT0990447. Turner

352 acknowledges an Australian Research Council Professorial Fellowship.

353

Figure captions

Figure 1. Map of the Kamchatka peninsula with the three main volcanic regions: the Central Kamchatka Depression (CKD), the Sredinny Ridge (SR) and the Eastern Volcanic Front (EVF). Triangles show the location of the main active volcanoes. Dashed curves show the depth of the subducting plate surface. Modified from (Dosseto et al. 2003).

Figure 2. U-series isotope compositions of Kamchatka lavas. Data from this study (diamonds) and (Dosseto et al. 2003) (triangles) were obtained on the same samples. Crosses: (Turner et al. 2007). In (a), significant differences are observed in a $(^{230}\text{Th}/^{232}\text{Th})$ vs. $(^{238}\text{U}/^{232}\text{Th})$ equiline diagram between this study and (Dosseto et al. 2003). This illustrates that this discrepancy is mostly the result of the poor quality of the Th data previously published. (b) The positive correlation between $(^{226}\text{Ra}/^{230}\text{Th})$ and $(^{231}\text{Pa}/^{235}\text{U})$ is still observed with the new measurements. Unlike in Dosseto et al. (2003) where the correlation is believed to represent mixing between melts derived from slab melting on the one hand, and mantle melting on the other hand, here it is proposed that magma differentiation produces this correlation, with a decrease in radioactive disequilibrium as differentiation takes place (see text for details). Compositions with the largest $(^{226}\text{Ra}/^{230}\text{Th})$ and $(^{231}\text{Pa}/^{235}\text{U})$ disequilibria are believed to represent those of a primitive melt produced from a metasomatised mantle: termed *HMB* in (Dosseto et al. 2003) (dark grey dot) or *primitive magma* (light grey area). The curve shows calculated compositions using a continuous flux melting model as in Dosseto et al. (2003). It is clear that following this model, the production of ^{226}Ra excess requires a decrease in the $(^{231}\text{Pa}/^{235}\text{U})$ which is not observed here. Errors are not shown for (Dosseto et al. 2003; Turner et al. 2007) data.

Figure 3. Differentiation model. It is possible to account for observed compositions using an assimilation/crystallisation model. However, only minimum crystallisation rates and maximum durations of differentiation can be inferred. Compositions can be explained by continuous crystallization at a minimum rate of $5 \times 10^{-6} \text{ g/g/yr}$ over a maximum duration of $\sim 6,000$ years. To account for high Th concentrations, assimilation is required and the ratio of assimilation to crystallisation rates needs to be between 0.1 and 0.3. Moreover, high U and Th concentrations in the assimilant are required in order to reproduce Sr/Th and $(^{231}\text{Pa}/^{235}\text{U})$ compositions simultaneously. In these diagrams, the dashed model curve is shown as a possible solution where the most evolved compositions are produced after up to 6,000 years of differentiation. The model curves are calculated with the following parameters: crystallisation rate = $5 \times 10^{-6} \text{ g/g/yr}$, assimilation/crystallisation rate ratio = 0 (plain curve) and 0.3 (dashed curve); parental magma composition: U = 0.1ppm, Th = 0.5ppm, Sr = 400ppm, $(^{226}\text{Ra}/^{230}\text{Th}) = 3.5$ and $(^{231}\text{Pa}/^{235}\text{U}) = 1.7$; assimilant composition: U = 60ppm, Th = 100ppm, Sr = 100ppm, $(^{226}\text{Ra}/^{230}\text{Th}) = 1$ and $(^{231}\text{Pa}/^{235}\text{U}) = 1$. Bulk partition coefficients are: $D_{\text{Th}} = 0.01$, $D_{\text{U}} = 0.05$, $D_{\text{Ra}} = 0.1$, $D_{\text{Pa}} = 0.0001$ and $D_{\text{Sr}} = 0.8$ (a high coefficient is taken for Ra in order to maximize the effect of crystal fractionation, however it has no effect on the calculated curves). Tick marks every 10,000 (plain curve) and 2,000 years (dashed curve). Dark grey diamonds: high-magnesia and high-alumina basalts; light grey diamonds: andesites. Errors are not shown for (Turner et al. 2007) data.

Figure 4. Dynamic melting of a source that has undergone recent gain of U and Ra (< 10 ka ago) via slab-derived fluid addition. Prior to melting, the metasomatized source has a $(^{230}\text{Th}/^{232}\text{Th})$ ratio of 1.3 (Turner et al. 1998) and $(^{238}\text{U}/^{232}\text{Th})$ ratios range between 1.8 and 2.1, reflecting the variable addition of fluids carrying U and Ra but no Th or Pa. This range of values is chosen such that the best fit to the data is produced. Bulk partition coefficients considered are: $D_{\text{Th}} = 0.00207$, $D_{\text{U}} = 0.00465$, $D_{\text{Pa}} = 0.000048$ and

$D_{Ra} = 0.000001$ (based on a source with 52% olivine, 20% orthopyroxene, 20% clinopyroxene and 8% garnet). Final degree of partial melting: $F=0.2$. Porosity: $\phi=0.001$. Melting rate = 2.83×10^{-4} g/g/yr and up/downwelling rate = 3 cm/yr. In (a), squares: this study, crosses: data from (Turner et al. 2007). In (b), diamonds: this study. Errors are not shown for (Turner et al. 2007) data.

Figure 5. Dynamic melting of a recently metasomatized source used to account for (a) Nicaragua and (b) Parinacota U-Th data. In (a), ($^{230}\text{Th}/^{232}\text{Th}$) of the source = 1.5 and ($^{238}\text{U}/^{232}\text{Th}$) ranges between 1.5 and 2.7. $D_{Th} = 0.00207$, $D_U = 0.00465$ (based on 52% ol, 20% opx, 20% cpx and 8% gt), $F=0.08$, $\phi=0.001$, melting rate = 2.64×10^{-4} g/g/yr and up/downwelling rate = 7 cm/yr. In (b), ($^{230}\text{Th}/^{232}\text{Th}$) of the source = 0.3, following sediment addition, and ($^{238}\text{U}/^{232}\text{Th}$) ranges between 0.3 and 0.9. $D_{Th} = 0.002069$, $D_U = 0.004651$ (based on 52% ol, 20% opx, 20% cpx and 8% gt), $F=0.08$, $\phi=0.001$, melting rate = 1.89×10^{-4} g/g/yr and up/downwelling rate = 5 cm/yr. Errors are not shown. Data are from (Reagan et al. 1994) for Nicaragua and (Bourdon et al. 2000) for Parinacota.

Figure 6. Dynamic melting under high f_{O_2} . The source has undergone addition of U during an old (>350 ka ago) fluid addition event (Conceição and Bonotto 2003), resulting in a range of ($^{230}\text{Th}/^{232}\text{Th}$) values prior to melting between 2.1 and 2.4. During melting, as a consequence of high oxygen fugacity, uranium is more incompatible than thorium ($D_U = D_{Th}/10$; see text for details). Other parameters are similar to previous models: $D_{Th} = 0.00207$ and $D_{Pa} = 0.000048$ (based on 52% ol, 20% opx, 20% cpx and 8% gt), $F=0.15$, $\phi=0.001$, melting rate = 3.54×10^{-4} g/g/yr and up/downwelling rate = 5 cm/yr. Shaded area in (a): data from (Turner et al. 2007). Errors are not shown for (Turner et al. 2007) data.

References

- Asmerom, Y., H. Cheng, R. Thomas, M. Hirschmann and R. L. Edwards (2000). "Melting of the Earth's lithospheric mantle inferred from protactinium- thorium-uranium isotopic data." *Nature* **406**(6793): 293-296.
- Avanzinelli, R., J. Prytulak, S. Skora, A. Heumann, G. Koetsier and T. Elliott (2012). "Combined ^{238}U – ^{230}Th and ^{235}U – ^{231}Pa constraints on the transport of slab-derived material beneath the Mariana Islands." *Geochimica et Cosmochimica Acta* **92**(0): 308-328.
- Beier, C., S. P. Turner, J. M. Sinton and J. B. Gill (2010). "Influence of subducted components on back-arc melting dynamics in the Manus Basin " *Geochemistry, Geophysics, Geosystems: G3* **11**.
- Blake, S. and N. Rogers (2005). "Magma differentiation rates from ($^{226}\text{Ra} / ^{230}\text{Th}$) and the size and power output of magma chambers." *Earth and Planetary Science Letters* **236**(3-4): 654-669.
- Bourdon, B. and T. Elliott (2010). Melt production in the mantle: constraints from U-series. *Timescales of magmatic processes: from core to atmosphere*. A. Dosseto, S. Turner and J. A. Van Orman. Oxford, Wiley-Blackwell.
- Bourdon, B., G. M. Henderson, C. C. Lundstrom and S. P. Turner (2003). *Uranium-series Geochemistry*. Washington, Geochemical Society - Mineralogical Society of America.
- Bourdon, B., S. Turner and A. Dosseto (2003). "Dehydration and partial melting in subduction zones: Constraints from U-series disequilibria." *Journal of Geophysical Research B: Solid Earth* **108**(6): ECV 3-1 - 3-19.
- Bourdon, B., G. Wörner and A. Zindler (2000). "U-series evidence for crustal involvement and magma residence times in the petrogenesis of Paríacota volcano, Chile." *Contrib. Mineral. Petrol.* **139**: 458-469.
- Conceição, F. T. and D. M. Bonotto (2003). *RADIOMETRIC CHARACTERIZATION OF SEDIMENTARY ROCKS AT CORUMBATAÍ RIVER BASIN, SÃO PAULO STATE, BRAZIL*. IV South American Symposium on Isotope Geology, Salvador, Brazil.
- Cooper, K. M. and M. R. Reid (2003). "Re-examination of crystal ages in recent Mount St. Helens lavas: implications for magma reservoir processes." *Earth and Planetary Science Letters* **213**(1-2): 149-167.
- Cooper, K. M., M. R. Reid, M. T. Murrell and D. A. Clague (2001). "Crystal and magma residence at Kilauea Volcano, Hawaii: ^{230}Th – ^{226}Ra dating of the 1955 east rift eruption." *Earth and Planetary Science Letters* **184**(3-4): 703-718.
- DeMets, C. R., G. Gordon, D. F. Argus and S. Stein (1990). "Current plate motions." *Geophys. J. Int.* **101**: 425-478.
- Dosseto, A., B. Bourdon, J.-L. Joron and B. Dupré (2003). "U-Th-Pa-Ra study of the Kamchatka arc: new constraints on the genesis of arc basalts." *Geochim. Cosmochim. Acta* **67**(15): 2857-2877.
- Dosseto, A. and S. Turner (2010). Magma cooling and differentiation – U-series isotopes. *Timescales of magmatic processes: from core to atmosphere*. A. Dosseto, S. Turner and J. A. Van Orman. Oxford, Wiley-Blackwell: 160-180.
- Dosseto, A., S. Turner and G. B. Douglas (2006). "Uranium-series isotopes in colloids and suspended sediments: timescale for sediment production and transport in the Murray-Darling River system." *Earth Planet. Sci. Lett.* **246**(3-4): 418-431.
- Dosseto, A., S. Turner and J. A. van Orman (2010). *Timescales of magmatic processes: from core to atmosphere*. Oxford, Wiley-Blackwell.

- Dosseto, A., S. P. Turner, M. Sandiford and J. Davidson (2008). Uranium-series isotope and thermal constraints on the rate and depth of silicic magma genesis. *Dynamics of crustal magma transfer, storage and differentiation*. C. Annen and G. Zellmer. London, Geological Society. **304**: 169-181.
- DuFrane, S. A., S. Turner, A. Dosseto and M. van Soest (2009). "Reappraisal of fluid and sediment contributions to Lesser Antilles magmas." *Chemical Geology* **265**(3-4): 272-278.
- Elliott, T., T. Plank, A. Zindler, W. White and B. Bourdon (1997). "Element transport from subducted slab to juvenile crust at the Mariana arc." *J. Geophys. Res.* **102**: 14991-15019.
- England, R. N. and H. L. Davies (1973). "Mineralogy of ultramafic cumulates and tectonites from eastern Papua." *Earth and Planetary Science Letters* **17**(2): 416-425.
- Hawkesworth, C. J., S. P. Turner, F. McDermott, D. W. Peate and P. van Calsteren (1997). "U-Th isotopes in arc magmas: implications for element transfer from the subducted crust." *Science* **276**: 551-555.
- Huang, F., L. Gao and C. C. Lundstrom (2008). "The effect of assimilation, fractional crystallization, and ageing on U-series disequilibria in subduction zone lavas." *Geochimica et Cosmochimica Acta* **72**(16): 4136-4145.
- Huang, F., C. C. Lundstrom, H. Sigurdsson and Z. Zhang (2011). "U-series disequilibria in Kick'em Jenny submarine volcano lavas: A new view of time-scales of magmatism in convergent margins." *Geochimica et Cosmochimica Acta* **75**(1): 195-212.
- Kersting, A. B. and R. J. Arculus (1995). "Pb isotope composition of Klyuchevskoy volcano, Kamchatka and North Pacific sediments: implications for magma genesis and crustal recycling in the Kamchatka arc." *Earth Planet. Sci. Lett.* **136**: 133-148.
- Landwehr, D., J. Blundy, E. M. Chamorro-Perez, E. Hill and B. Wood (2001). "U-series disequilibria generated by partial melting of spinel lherzolite." *Earth and Planetary Science Letters* **188**(3-4): 329-348.
- Lundstrom, C. C., H. F. Shaw, F. J. Ryerson, D. L. Phinney, J. B. Gill and Q. Williams (1994). "Compositional controls on the partitioning of U, Th, Ba, Pb, Sr and Zr between clinopyroxene and haplobasaltic melts: implications for uranium series disequilibria in basalts." *Earth Planet. Sci. Lett.* **128**: 407-423.
- McKenzie, D. (1985). "²³⁰Th-²³⁸U disequilibrium and the melting processes beneath ridge axes." *Earth Planet. Sci. Lett.* **72**: 149-157.
- Melekestsev, I., A. P. Khrenov and N. N. Kozhemyaka (1991). Tectonic position and general description of volcanoes of Northern group and Sredinny range. *Active volcanoes of Kamchatka*. A. o. Sciences. Moscou. **n°1. partie II (avant-propos)**: 79-97.
- Miyashiro, A. (1974). "Volcanic rock series in island arcs and active continental margins." *Am. J. Sci.* **274**: 321-355.
- Peate, D. W. (2005). "U series disequilibria: Insights into mantle melting and the timescales of magma differentiation." *Reviews of Geophysics* **43**(1).
- Petford, N., M. A. Koenders and S. Turner (2008). "Channelized melt flow in downwelling mantle: Implications for ²²⁶Ra-²¹⁰Pb disequilibria in arc magmas." *Journal of Geophysical Research* **113**(B11).
- Pineau, F., M. P. Semet, N. Grassineau, V. M. Okrugin and M. Javoy (1999). "The genesis of the stable isotope (O,H) record in arc magmas: the Kamtchatka's case." *Chem. Geol.* **153**: 93-124.
- Price, R. C., et al. (2007). "U-Th-Ra fractionation during crustal-level andesite formation at Ruapehu volcano, New Zealand." *Chemical Geology* **244**(3-4): 437-451.
- Reagan, M. K., J. D. Morris, E. A. Herstrom and M. T. Murrell (1994). "Uranium series and beryllium isotope evidence for an extended history of subduction modification of the mantle below Nicaragua." *Geochim. Cosmochim. Acta* **58**(19): 4199-4212.

- Reagan, M. K., F. J. Tepley III, J. B. Gill, M. Wortel and J. Garrison (2006). "Timescales of degassing and crystallization implied by ^{210}Po - ^{210}Pb - ^{226}Ra disequilibria for andesitic lavas erupted from Arenal volcano." *Journal of Volcanology and Geothermal Research* **157**(1-3): 135-146.
- Rogers, N. W., P. J. Evans, S. Blake, S. C. Scott and C. J. Hawkesworth (2004). "Rates and timescales of fractional crystallization from ^{238}U - ^{230}Th - ^{226}Ra disequilibria in trachyte lavas from Longonot volcano, Kenya." *Journal of Petrology* **45**(9): 1747-1776.
- Rubin, K. H. and J. D. Macdougall (1988). " ^{226}Ra excesses in mid-ocean-ridge basalts and mantle melting." *Nature* **335**(6186): 158-161.
- Rubin, K. H., I. Van Der Zander, M. C. Smith and E. C. Bergmanis (2005). "Minimum speed limit for ocean ridge magmatism from ^{210}Pb - ^{226}Ra - ^{230}Th disequilibria." *Nature* **437**(7058): 534-538.
- Sigmarsson, O. (1996). "Short magma chamber residence time at an Icelandic volcano inferred from U-series disequilibria." *Nature* **382**(6590): 440-442.
- Sigmarsson, O., M. Condomines and P. Bachèlery (2005). "Magma residence time beneath the Piton de la Fournaise Volcano, Reunion Island, from U-series disequilibria." *Earth and Planetary Science Letters* **234**(1-2): 223-234.
- Sims, K. W. W., D. J. DePaolo, M. T. Murell, W. S. Baldrige, S. Goldstein, D. Clague and M. Jull (1999). "Porosity of the melting zone and variations in the solid mantle upwelling rate beneath Hawaii: Inferences from ^{238}U - ^{230}Th - ^{226}Ra and ^{235}U - ^{231}Pa disequilibria." *Geochim. Cosmochim. Acta* **63**(23/24): 4119-4138.
- Sims, K. W. W., D. J. DePaolo, M. T. Murell, W. S. Baldrige, S. J. Goldstein and D. A. Clague (1995). "Mechanisms of magma generation beneath Hawaii and mid-ocean ridges: Uranium/thorium and samarium/neodymium isotopic evidence." *Science* **267**: 508-512.
- Sims, K. W. W., et al. (2008). "An Inter-Laboratory Assessment of the Thorium Isotopic Composition of Synthetic and Rock Reference Materials." *Geostandards and Geoanalytical Research* **32**(1): 65-91.
- Sims, K. W. W., et al. (2002). "Chemical and isotopic constraints on the generation and transport of magma beneath the East Pacific Rise." *Geochimica et Cosmochimica Acta* **66**(19): 3481-3504.
- Sims, K. W. W., et al. (2008). " ^{238}U - ^{230}Th - ^{226}Ra - ^{210}Pb - ^{210}Po , ^{232}Th - ^{228}Ra , and ^{235}U - ^{231}Pa constraints on the ages and petrogenesis of Vailulu'u and Malumalu Lavas, Samoa." *Geochemistry Geophysics Geosystems* **9**(4).
- Spiegelman, M. (2000). "UserCalc: a web-based uranium series calculator for magma migration problems." *Geochem. Geophys. Geosyst.* **1**.
- Spiegelman, M. and T. Elliott (1993). "Consequences of melt transport for uranium series disequilibrium in young lavas." *Earth Planet. Sci. Lett.* **118**: 1-20.
- Stracke, A., B. Bourdon and D. McKenzie (2006). "Melt extraction in the Earth's mantle: Constraints from U-Th-Pa-Ra studies in oceanic basalts." *Earth and Planetary Science Letters* **244**(1-2): 97-112.
- Straub, S. M., A. Gomez-Tuena, F. M. Stuart, G. F. Zellmer, R. Espinasa-Perena, Y. Cai and Y. Iizuka (2011). "Formation of hybrid arc andesites beneath thick continental crust." *Earth and Planetary Science Letters* **303**(3-4): 337-347.
- Tegner, C., J. R. Wilson and B. Robins (2005). "Crustal assimilation in basalt and jotunite: Constraints from layered intrusions." *Lithos* **83**(3-4): 299-316.
- Tepley III, F. J., C. C. Lundstrom, J. B. Gill and R. W. Williams (2006). "U-Th-Ra disequilibria and the time scale of fluid transfer and andesite differentiation at Arenal volcano, Costa Rica (1968-2003)." *Journal of Volcanology and Geothermal Research* **157**: 147-165.
- Thomas, R. B., M. M. Hirschmann, H. Cheng, M. K. Reagan and R. L. Edwards (2002). " $^{231}\text{Pa}/^{235}\text{U}$ - $^{230}\text{Th}/^{238}\text{U}$ of young mafic volcanic rocks from Nicaragua and Costa Rica and the influence of

- flux melting on U-series systematics of arc lavas." *Geochimica et Cosmochimica Acta* **66**(24): 4287-4309.
- Touboul, M., B. Bourdon, B. Villemant, G. Boudon and J.-L. Joron (2007). "²³⁸U-²³⁰Th-²²⁶Ra disequilibria in andesitic lavas of the last magmatic eruption of Guadeloupe Soufriere, french Antilles: Processes and timescales of magma differentiation." *Chemical Geology* **246**(3-4): 181-206.
- Turner, S., B. Bourdon and J. Gill (2003). Insights into Magma Genesis at Convergent Margins from U-series Isotopes. *Uranium-series Geochemistry*. B. Bourdon, G. M. Henderson, C. C. Lundstrom and S. P. Turner. Washington, Geochemical Society - Mineralogical Society of America. **52**: 255-315.
- Turner, S., B. Bourdon, C. Hawkesworth and P. Evans (2000). "²²⁶Ra-²³⁰Th evidence for multiple dehydration events, rapid melt ascent and time scales of differentiation beneath the Tonga-Kermadec island arc." *Earth Planet. Sci. Lett.* **179**: 581-593.
- Turner, S., P. Evans and C. Hawkesworth (2001). "Ultrafast source-to-surface movement of melt at island arcs from ²²⁶Ra-²³⁰Th systematics." *Science* **292**: 1363-1366.
- Turner, S. and C. Hawkesworth (1997). "Constraints on flux rates and mantle dynamics beneath island arcs from Tonga-Kermadec lava geochemistry." *Nature* **389**: 568-573.
- Turner, S., F. McDermott, C. Hawkesworth and P. Kepezhinskis (1998). "A U-series study of lavas from Kamchatka and the Aleutians: constraints on source composition and melting processes." *Contrib. Mineral. Petrol.* **133**: 217-234.
- Turner, S., D. W. Peate, C. J. Hawkesworth, S. M. Eggins and A. J. Crawford (1999). "Two mantle domains and the time scales of fluid transfer beneath the Vanuatu arc." *Geology* **27**(11): 963-966.
- Turner, S., M. Regelous, C. Hawkesworth and K. Rostami (2005). "Partial melting processes above subducting plates: Constraints from ²³¹Pa-²³⁵U disequilibria." *Geochimica et Cosmochimica Acta* **70**: 480-503.
- Turner, S., K. W. W. Sims, M. Reagan and C. Cook (2007). "A ²¹⁰Pb-²²⁶Ra-²³⁰Th-²³⁸U study of Klyuchevskoy and Bezymianny volcanoes, Kamchatka." *Geochimica et Cosmochimica Acta* **71**(19): 4771-4785.
- Zou, H. and A. Zindler (2000). "Theoretical studies of ²³⁸U-²³⁰Th-²²⁶Ra and ²³⁵U-²³¹Pa disequilibria in young lavas produced by mantle melting." *Geochimica et Cosmochimica Acta* **64**(10): 1809-1817.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

(a)

$(^{230}\text{Th}/^{232}\text{Th})$

(b)

$(^{230}\text{Th}/^{232}\text{Th})$

Figure 6

Table 1. Geochemical data for Kamchatka samples

Sample name	Eruption date	Th (ppm)	U (ppm)	²²⁶ Ra (fg/g)	Sr (ppm)	(²³⁴ U/ ²³⁸ U)	(²³⁰ Th/ ²³⁸ U)	(²²⁶ Ra/ ²³⁰ Th)	(²³¹ Pa/ ²³⁵ U)	(²³⁸ U/ ²³² Th)	(²³⁰ Th/ ²³² Th)	Sr/Th
<i>Tolbachik</i>												
PT30-4/1976	August 1976	1.687	1.035	676.8	358	0.992	0.982	1.97	1.69	1.861	1.829	212
PT33-1/82	Holocene	0.884	0.557	188.6	285	0.996	0.986	1.02	1.13	1.914	1.887	322
PT33-1/82-R		0.834	0.524	190.1		0.998	0.989	1.09	1.20	1.908	1.887	
T1475/1975	30 July 1975	0.534	0.333	346.0	461	0.995	0.970	3.17	1.69	1.892	1.835	863
<i>Bezymianny</i>												
M-1564-1	post-1956 cycle	1.298	0.917	285.7	323	0.993	0.939	0.98	1.00	2.143	2.013	249
B82	1982	1.126	0.794	342.9	353	0.998	0.935	1.37	1.42	2.141	2.003	313
B8/4	1986	0.735	0.517	326.1	323	0.990	0.938	1.99	2.08	2.136	2.003	440
<i>Kliuchevskoi</i>												
AXO3/84	1984	0.736	0.481	261.5	376	0.982	0.940	1.71	1.26	1.983	1.864	511
AXO3/84-R		0.725	0.477	254.1		0.975	0.937	1.68	1.27	1.997	1.871	
K56P66	1966	0.722	0.473	264.5	362	0.999	0.962	1.72	1.37	1.991	1.916	502
<i>Gravimetric standards</i>												
BCR-2 (n=4)		5.628	1.612			0.991	0.999			1.001	0.868	
TML-3 (n=11 for U-Th; n=1 for Ra)		29.69	10.59	3550		1.003	0.994	0.996		1.006	1.067	
Relative 2 standard error (%)		0.0	0.0	0.0		0.0	0.0	0.0	0.0	0.0	0.0	

Relative 2 standard errors are calculated using the worst sample replicate analysis (PT33-1/82 or AXO3/84)