

$^{226}\text{Ra}/^{230}\text{Th}$ excess generated in the lower crust: Implications for magma transport and storage time scales: Comment and Reply

COMMENT doi:10.1130/G22667.1

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Dufek and Cooper (2005; hereafter DC) present an interesting and provocative paper on the origins of ^{226}Ra excess in arc lavas. They develop an incongruent continuous melting model to argue that ^{226}Ra excess can be created by partial melting in the lower crust. However, despite the novelty of their model, the conclusions of DC are questionable for three reasons.

Firstly, one of the reasons for developing the continuous melting model is that DC argue that melt can segregate at low melt fractions (1%–10%). At such low melt fractions, accessory phases such as zircon and allanite are saturated in crustal melt because of their relatively low solubility. Although the abundance of these elements may be low, they control U-series geochemistry because their partition coefficients are several orders of magnitude higher than those of major silicate phases (e.g., Berlo et al., 2004). This implies that we would expect U-series disequilibria at small melt fractions to be even higher, exceeding observations.

Secondly, partition coefficients depend on pressure, temperature, and composition, and therefore must be carefully matched to the problem under investigation. Because the partition coefficients of Ra, Th, and U are all very low, small variations can have a large impact on the calculated disequilibria. DC employ a compilation from a large range of systems (amphibolite to lherzolite) and a range in P - T conditions (mantle to crust). The problem of partition coefficients is particularly acute in the case of incongruent melting if product phases have vastly different partition coefficients for parent and daughter nuclides. During non-modal melting of amphibolite, cpx is a product phase. In the calculations of DC, cpx is assigned D_U and D_{Th} close to unity, while D_{Ra} is very small. Consequently, the transfer of U and Th to the melt is held back relative to Ra, and large Ra excesses develop. This can be anticipated without the development of a complex mathematical model. For example, using the DC values for D_{Th} and D_U , the simple batch model of Berlo et al. (2004) generates ^{226}Ra excesses up to 4 at 20% of melting. DC's choice of D 's for cpx comes largely from the study of Barth et al. (2002), who report $D_{Th} = 0.88 \pm 0.22$. This high value for D_{Th} is wholly at odds with what is known about mineral-melt partitioning (Fig. 1). We contend that near-unity D 's for U and Th are inappropriate for modeling crustal melting. Appropriate D 's for U and Th would be a factor of ~20 lower (e.g., Blundy and Wood, 2003). Such a low value of D_{Th} would not result in significant ^{226}Ra excess, even with the model of DC.

Thirdly, implicit in the paper by DC is the notion that their model is applicable to most arc rocks. As DC note, "amphibolite melts will dominate the trace element budget of mixtures even when they represent a small mass fraction" (Dufek and Cooper, 2005, p. 835). Melts formed in equilibrium with residual garnet carry distinctive trace element signatures that can be resolved from those of primary arc liquids. Log-normalized diagrams can hide much important detail, but inspection of Figure 2A in DC (2005) shows that their modeled lower crustal melts increase ratios

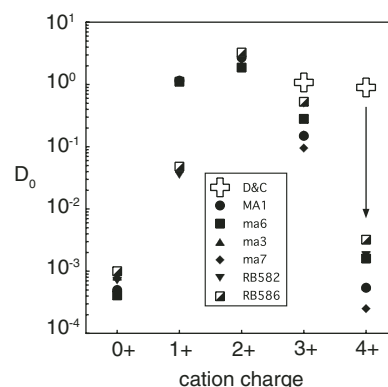


Figure 1. Plot of strain-compensated partition coefficient (D_0) versus cationic charge (Zc) for cpx-melt experiments of Brooker et al. (2003), which cover a wide range in pressure (0.1–8.1 GPa) and temperature (1200–1660 °C), and data used by Dufek and Cooper (2005). The high value of D_0 for 4+ cations (U and Th) in the data set of Dufek and Cooper are inconsistent.

such as Sr/Y and La/Yb by factors of ~8 and ~13, respectively, relative to the starting composition. Thus, the mixed melt plotted in DC's Figure 2B has a Sr/Y ratio of ~62 and a La/Yb ratio of ~6.7, in marked contrast to the average arc rock Sr/Y and La/Yb ratios of 19 and 4.3 (calculated using the data compilation from Turner et al. 2003, not filtered for SiO_2). Moreover, there is no positive correlation between $^{226}\text{Ra}/^{230}\text{Th}$ and either Sr/Y or La/Yb, as would be predicted if the DC model had general application. Indeed, the highest ^{226}Ra excesses have been observed in those rocks with the lowest La/Yb ratios. Rather, elevated Sr/Y and La/Yb ratios are characteristic of quite rare arc rocks commonly referred to as adakites. It has long been argued that adakites are formed by partial melting of amphibolite, either in the lower crust or in the subducting slab. Thus, the DC model may be highly applicable to adakites, but far less so to the vast majority of arc rocks on which hypotheses for rapid melt ascent and differentiation have been based.

In summary, we agree with DC that lower crustal melting may be important in modifying mantle-derived U-series signatures in some arc lavas, especially those erupted through thick continental crust. However, such conclusions are based on the choice of partition coefficients rather than the choice of melting model. For the bulk of island arc lavas, we argue that the effect of interaction with the lower crust will usually be to reduce the extent of mantle derived disequilibria. Thus, the time scales for magma ascent and differentiation inferred from U-series isotope measurements can be treated as maxima.

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REPLY doi: 10.1130/G23073.1

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We welcome the comments by Berlo et al. (hereafter BTBH) and thank them for the opportunity to further discuss lower crustal melting processes in arc settings. Our intention in Dufek and Cooper (2005) was not to explain the U-series disequilibria in all arc magmas through a single model. Instead, because the composition of many arc magmas is the result of polybaric processes, we explored whether U-series disequilibria measured at the surface may reflect some contribution from lower crustal melting where the crust is >30 km thick. We disagree with BTBH that the form of the melting model is insignificant, considering field and experimental evidence for melt segregation at low melt fractions. Here we discuss the three comments made by BTBH and show that significant ^{226}Ra - ^{230}Th disequilibria can be produced during continuous, incongruent melting, even using the partition coefficients favored by BTBH.

It is well established that models that explicitly consider the duration of melting with low residual porosity (~0.1%) are required to explain observed U-series disequilibria for mid-oceanic-ridge basalts (e.g., Lundstrom, 2003). However, BTBH (2004; and in their Comment) argue that partial melts of lower crust are unlikely to segregate at melt fractions below 20%–625%. As we discussed in our paper, use of a time-dependent melting model is consistent with experimental and field evidence of hydrous melt segregation at low melt fraction ($\leq 10\%$), often during synchronous deformation (Brown, 2005). Furthermore, while we did advocate small critical porosities relative to batch melting, this is significantly different from stating that we examined only small total melt fractions (i.e., limited progress in the dehydration reaction); in fact, our Figure 1 shows $(^{226}\text{Ra})/(^{230}\text{Th}) > 3$ at up to 0.25 extracted melt fraction, provided that the melting rate is lower than $\sim 10^{-2}$ kg/m³/yr. We agree that zircon, if present in the residue or fractionated from the ascending liquid, will likely act to increase $(^{226}\text{Ra})/(^{230}\text{Th})$. However, this process would only produce excessively large $(^{226}\text{Ra})/(^{230}\text{Th})$ ratios at the surface if 1) lavas at the surface sample these melts directly, without mixing with other melts, and 2) the magma does not stall for any appreciable time in transit. We suggested in our paper that erupted lavas may reflect mixtures of mantle-derived magmas with lower-crustal melts, and the larger Ra excesses predicted in the presence of residual accessory phases would relax the necessary ascent rates and would increase the range of applicability of this model.

BTBH's second point relates to the partition coefficients used in our model. We fully agree that pressure, temperature, and composition must be taken into account when modeling melting processes. We used partition coefficients from the literature that reported *P-T-X* most like the phases in the Wolf and Wyllie (1994) amphibolite melting experiments that we used as a model melting reaction. We were not able to evaluate the appropriateness of the partition coefficients used by Berlo et al. (2004) because data on the experimental compositions were not available. Here we examine the sensitivity of the continuous melting model to the choice of partition coefficients by performing calculations using those suggested by

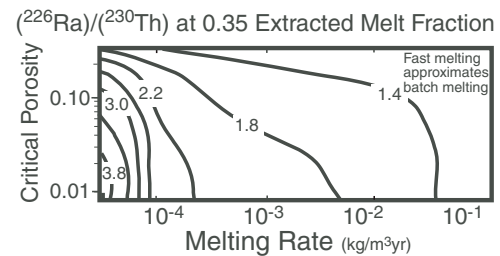


Figure 1. Time-dependent incongruent melting model using partition coefficients suggested by Berlo et al. (2004).

Berlo et al. (2004), and at similarly high melt fraction (0.35) as their batch melting calculations (Fig. 1). Batch melting is most closely approximated by very fast melting, which produces almost no ^{226}Ra excess. However, at slower and more geologically relevant melting rates, ^{226}Ra excesses develop that are consistent with many continental arc magmas, even with the partition coefficients favored by BTBH. Conduction and even comparatively fast processes such as gas sparging are unlikely to generate average melting rates greater than 10^{-3} kg/m³/yr (Bachmann and Bergantz, 2006; Dufek and Bergantz, 2005).

We proposed using the trace element signature of the incongruent melting process to identify magmas where the U-series disequilibria have been affected by lower-crustal melting. We agree with BTBH, that based on trace-element criteria, many island arc basalts erupted through a thin (< 30 km) crust are unlikely to have large crustal contributions to their U-series disequilibria. However, there are many settings (especially in continental arcs) where this process remains viable. We reiterate that both the choice of a primitive island arc basalt starting composition and the 50:50 mixing calculation of crustal and mantle melts presented in our paper were intended to be illustrative rather than definitive. Further, provided that melting is slow enough that in-growth effects are important, ^{226}Ra excess can be preserved to high melt fraction over a range of Sr/Y and La/Yb.

In summary, we agree with BTBH that rapid batch melting in the crust is unlikely to produce significant ^{226}Ra - ^{230}Th disequilibria. However, field and experimental evidence, combined with numerical models of melting rates within the crust, suggest that melt extraction at relatively small porosities over an extended period of time is likely. Time-dependent melting in the lower crust, especially in the presence of residual garnet, can produce Ra excesses and trace element signatures similar to those observed in a number of arc settings, and this needs to be considered when quantifying magma ascent and storage time scales in these settings.

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