# Diapirs as the source of the sediment signature in arc lavas

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Island arc lavas, erupted above subduction zones, commonly contain a geochemical component derived from partial melting of subducted sediment. It is debated whether this sediment melt signature, with enriched trace element concentrations and isotope ratios, forms at relatively low or high temperatures. Here we compile and analyse the geochemistry of metamorphosed sedimentary rocks that have been exposed to pressures between 2.7 and 5 GPa during subduction at a range of locations worldwide. We find that the trace elements that form the sediment melt signature are retained in the sediments until the rocks have experienced temperatures exceeding 1,050 °C. According to thermal models, these temperatures are much higher than those at the surface of subducted slabs at similar pressures. This implies that the sediment melt signature cannot form at the slab surface. Using instability calculations, we show that subducted sediments detach from the downgoing slab at temperatures of 500-850 °C to form buoyant diapirs. The diapirs rise through the overlying hot mantle wedge, where temperatures exceed 1,050 °C, undergo dehydration melting, and release the trace elements that later form the sediment melt signature in the erupted lavas. We conclude that sediment diapirism may reduce the transport of trace elements and volatiles such as CO<sub>2</sub> into the deep mantle.

B a, Th, Be, Pb, and light rare-earth elements (REE) are enriched in partial melts of metasediment, but many of these elements are relatively immobile in aqueous fluids<sup>1</sup>. The enrichment of these elements, and their correlation with the flux and composition of subducted sediments, has been interpreted to reflect a 'sediment melt' signature in arc lavas<sup>2-4</sup>. Subduction zone thermal models that incorporate temperature- and stress-dependent viscosity<sup>5-8</sup> produce slab-top temperatures above the fluid-saturated sediment solidus (>600–700 °C at ≤3 GPa; refs 9,10), and H<sub>2</sub>O/Ce and H<sub>2</sub>O/K ratios in melt inclusions from arc lavas are consistent with fluid fluxed melting of sediments at 750° to more than 950 °C beneath several global subduction systems<sup>11–13</sup>. Alternatively, it has been suggested that subducting sediment detaches as solid-state diapirs, and melts at higher temperatures as it ascends through the mantle wedge<sup>14–17</sup>.

The key to distinguishing between these models is determining the conditions under which sediment melting occurs in subduction zones. A major challenge in evaluating these conditions is the difficulty in sampling the residues of sediment melts. Most studies of rocks exhumed from subduction zones focus on either (1) basaltic compositions analogous to subducted oceanic crust or (2) unusual metasediments that attained high temperatures at relatively low peak pressures, different from even the hottest slab-top geotherms. Mafic ultrahigh-pressure (UHP) rocks subducted to pressure-temperature conditions above the fluid-saturated basalt solidus show MORB-like concentrations in fluid-immobile elements<sup>18</sup>, such as Th and La. This suggests that either melting was suppressed in these rocks because of low H<sub>2</sub>O activity or that minor phases retained Th, La, and other light REE in the solid residue. Similarly, metasediments from Santa Catalina Island<sup>19</sup> show only minor depletion of Th, La, Pb and Sr. However, these samples attained peak pressure-temperature conditions of 600 °C and 1-1.2 GPa, a higher temperature at a

given pressure than any steady state subduction geotherm, yet below the fluid-saturated sediment solidus. In another example, high-pressure and UHP metasediments from the western Alps that reached peak temperatures  $\leq 630 \,^{\circ}$ C along a plausible subduction geotherm show little sign of depletion of fluid-immobile elements compared to average shale<sup>20</sup>.

## Characterizing the conditions of sediment melting

To characterize the residues of sediment melting, we analysed and compiled other data on high- to UHP rocks (peak pressures mostly between 2.7 and 5 GPa; hereafter loosely termed UHP) associated with subduction systems to assess whether these rocks lost Th, Pb, and other fluid-immobile trace elements. To isolate pelitic sediment protoliths from igneous compositions we considered only peraluminous UHP compositions (see Supplementary Information for data sources and the method for identifying UHP metapelites). Comparing these data to a compilation of peraluminous shale compositions representative of the incoming sediment at subduction zones (see Supplementary Information for data sources), we find that UHP metapelites with peak temperatures of 700°-1,050 °C have compositions that lie within the compositional range of shale (Fig. 1a). This indicates that the UHP rocks are indeed representative of peraluminous subducting, pelitic sediments. However, many UHP metapelites that attained temperatures >1,050 °C show depletions in highly incompatible elements, including Th, Sr, Pb, and Nd, as expected for the residues of melting that could result in extensive recycling of the 'sediment component' from subducting metasediments to arc magmas (Fig. 1 and Supplementary Figures). Even among our samples that reached >1,050 °C, some are not depleted, suggesting that yet higher degrees of melting and/or more efficient melt extraction are required to fully exhaust the minor phases that host key trace elements in the solid residue.

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**Figure 1** | **Comparison of pelitic UHP metasediment compositions to average shale, greywacke and loess compositions. a**, Trace element diagram for pelitic UHP metasediments. 1 $\sigma$  envelope for average shale, greywacke, and loess is shown as blue field with black outlines. **b**-**e**, Trace element concentrations in pelitic UHP metasediments (red diamonds, see Supplementary Information for error estimates) as a function of peak metamorphic temperature compared to our average peraluminous shale and greywacke composition (black circle at arbitrary temperature of 600 °C), our compiled peraluminous shale, greywacke, and loess compositions (small grey squares, full data range denoted by light grey shaded region), and 1 $\sigma$  for our average shale and greywacke compositions of residues of partial melting of pelitic red clay<sup>23</sup> (blue squares with cross). (See Supplementary Information for data sources.)

For example, we observe phengite within garnet cores in many of our samples at temperatures  $\geq 1,000$  °C, indicating that phengite remained stable up to peak metamorphic conditions—as observed experimentally by Hermann and Green (ref. 21), who report residual phengite at 1,000 °C at 4 and 4.5 GPa, and as in Schmidt *et al.* (ref. 11), who infer that fluid-undersaturated, dehydration melting of phengite occurs from 950 °C at 3 GPa to 1,150 °C at 5 GPa (their Figure 11). We note that some fluid-saturated<sup>22</sup> experiments show lower temperatures for phengite breakdown, however, our results indicate that dehydration melting is a better approximation to the conditions in natural UHP metamorphism.

Our results are consistent with experimental studies of metasediments and metabasalts<sup>12,23,24</sup>, which show little depletion

in key trace elements during dehydration melting at  $600^{\circ}-1,000^{\circ}C$  and 2–5 GPa (Fig. 1b–e). The lack of depletion is probably due to the stability of minor phases (for example, phengite, allanite, apatite, monazite, and zircon) that are stable above the fluid-saturated solidus at UHP conditions and retain these key elements<sup>24–26</sup>.

Overall, the compiled UHP data imply that recycling of elements such as Th, La, Nd and Pb from metasediments is inefficient if slab-top temperatures are <1,000 °C in the region of arc magma genesis. Although the parameters involved in the mass balance are uncertain, subducting fluxes of Th, U, K, La and Nd, combined with primitive magmatic concentrations of these elements<sup>2</sup> and estimates for magmatic fluxes in arcs, generally yield recycling rates of ~100% (see Supplementary Information for these calculations).

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**Figure 2** | **Density of subducted sediments at UHP conditions.** Density calculated at 700 °C and **a**, 1 GPa and **b**, 3 GPa as a function of SiO<sub>2</sub> content for compiled UHP metasedimentary compositions (dark blue squares), our estimates of average shale and greywacke, average loess, and the average of prior shale compositional averages (light blue circles), and subducting sediment delineated by lithology (yellow diamonds<sup>30</sup>) and location (green triangles<sup>30</sup>). Average lower continental and arc lower crust<sup>50</sup> are shown with red and grey stars, respectively. Black and grey horizontal lines show densities calculated for harzburgite<sup>14</sup> and pyrolite<sup>35</sup>, respectively. Relative to the mantle wedge almost all sediment compositions are buoyant at 700 °C and pressures <3 GPa.

Thus, the apparent efficiency of recycling together with UHP metapelite compositions suggests that the sediment-melt signature in arcs is generated at >1,000 °C—significantly above the fluid-saturated sediment solidus. Temperatures greater than 1,000 °C are not reached on the slab top at pressures less than 5 GPa in even the hottest subduction-zone thermal models. Thus, we argue that the most likely location for the UHP metapelites to have reached temperatures >1,000 °C beneath volcanic arcs is within the mantle wedge. A possible example of this process is recorded by a peraluminous xenolith with oxygen isotopes indicative of a metasedimentary protolith from South Africa, which records peak metamorphic conditions of 1, 200–1, 300 °C and 4–5 GPa (that is, mantle wedge rather than subduction zone conditions)<sup>27</sup>.

## Timescale for the formation of sediment diapirs

Numerical modelling studies have shown the potential for subducting sediments to rise buoyantly into the mantle wedge<sup>17,28,29</sup>. However, these studies focused on systems in which the sediments are entrained in buoyant, hydrated mantle diapirs<sup>17,28</sup>, or on very thick (>2 km) sediment layers<sup>29</sup>, not representative of the thickness of sediment subducting to depth at modern arcs. Moreover, no studies have yet presented formal non-Newtonian scaling for sediment diapirs as a function of such key parameters as sediment thickness and density—preventing quantitative constraints on where and when sediment diapirs may form during subduction. Here we present a new, non-Newtonian scaling analysis for subducting sediments, to assess the timescale and depth at which a sediment layer may become unstable even in the absence of other sources of buoyancy (for example, a hydrated mantle layer).

To evaluate the intrinsic buoyancy of subducting sediments, we first calculated densities for the compiled UHP metapelites and averages of pelitic sediment compositions (ref. 30; Supplementary Information) and compared them to the density of the overlying mantle wedge (see Methods section). At 700 °C and pressures  $\leq$ 3 GPa, almost all UHP rocks and sediment averages are buoyant with respect to the mantle wedge (Fig. 2). Further, for both warm (Cascadia) and cold (Izu-Bonin) slab-top geotherms<sup>8</sup> the density contrast between the overlying mantle and the average UHP metapelite is as much as  $-200 \text{ kg m}^{-3}$  for all pressures up to 6 GPa (Fig. 3).

The timescale over which instabilities grow in a buoyant sediment layer is related to the relative viscosities of the sediment and overlying mantle, the viscous decay length in the mantle, and the buoyancy and thickness of the sediment layer<sup>31-35</sup>. We calculated instability times assuming a wet olivine rheology for the mantle wedge<sup>36</sup> that is  $100 \times$  more viscous than the underlying



**Figure 3** | Density of the average UHP metasediment along typical subduction zone geotherms. a, Density contrast between the average UHP metasedimentary composition and mantle wedge harzburgite<sup>14</sup> as a function of temperature and pressure. Thick red and black lines show slab-top geotherms for Cascadia and Izu-Bonin, respectively<sup>8</sup>. **b**, Density contrast as a function of pressure along the Cascadia (red) and Izu-Bonin (black) slab-top geotherms. Note that the density contrast is  $\leq -200 \text{ kg m}^{-3}$  for all pressures <6 GPa.

sediment layer (Fig. 4, see Methods section). This viscosity contrast is appropriate for wet quartz<sup>37</sup> at temperatures between 600° and 800 °C. For a 500 m-thick sediment layer with a density contrast of  $-200 \text{ kg m}^{-3}$  relative to the overlying mantle, the timescale for instability formation is <1 Myr and <10 kyr at temperatures of 700 °C and 800 °C, respectively (Fig. 4a and grey region in Fig. 4b). Moreover, any sediment layer thicker than ~100 m will become unstable on timescales shorter than 1 Myr at temperatures  $\leq$ 1,000 °C (Fig. 4a).

The conditions for diapir formation in a given subduction zone can be determined by integrating the instability time along the slabtop pressure–temperature–time (P-T-t) path until the amplitude of the instability exceeds the initial thickness of the sediment layer. To illustrate this approach, the P-T conditions at which diapirs form in Cascadia and Izu–Bonin were calculated assuming a sediment thickness of 500 m (stars on the black and red curves in Fig. 4b). In both cases, diapirs form between ~675°–750 °C and



Figure 4 | Calculated timescale for the initiation of a sediment diapir. a, Instability time versus temperature and sediment layer thickness for a density contrast of −200 kg m<sup>-3</sup>, background strain-rate of 10<sup>-16</sup> s<sup>-1</sup>, and initial perturbation amplitude 33% of the layer thickness. For typical subduction rates, diapirs form between 2 and 4 GPa if the instability time is ≤1 Myr (thick black contour). **b**, Location of diapir formation (stars) on slab-top geotherms for Cascadia (red) and Izu-Bonin (black) assuming a sediment thickness of 500 m. Grey region shows instability times for background strain-rates ranging from 10<sup>-14</sup> to 10<sup>-18</sup> s<sup>-1</sup>. Diapirs are predicted to form at 685 °C and ~2.3 GPa in Cascadia and 730 °C and 3.2 GPa in Izu-Bonin.

2.2–3.0 GPa, with a slightly greater formation depth in Izu–Bonin due to the colder incoming slab.

Based on slab-top geotherms<sup>8</sup> and the estimated thickness of the downgoing sediment layer<sup>38,39</sup>, we explored the conditions of diapir formation for 17 different subduction systems (Fig. 5). For all subduction systems, diapirs form between 500° and 850°C (Fig. 5a), with higher temperatures corresponding to thinner sediment layers because instability growth rates scale positively with layer thickness<sup>31,34,35</sup>. These temperatures are near the fluidsaturated sediment solidus, but significantly below the ~1,050°C required to deplete metasediments in Th, Sr, Pb and Nd (Fig. 1). Thus, we conclude that diapirs of buoyant metasediment detach from the downgoing slab and rise buoyantly into the mantle wedge, where they are heated to temperatures >1,000–1,100°C.



**Figure 5** | Summary of conditions for sediment diapir formation in global subduction zones. a, Diapir initiation temperature versus sediment layer thickness<sup>30</sup>, and **b**, diapir initiation depth versus subarc slab depth (as compiled in ref. 8) calculated for 17 slab-top geotherms<sup>8</sup>. Subducting sediment layer thicknesses are corrected for compaction to a density of 2,800 kg m<sup>-3</sup>. Numbers (in order of increasing slab thermal parameter) correspond to subduction zones: 1—Cascadia, 2—Nankai, 3—Mexico, 4—Colombia-Ecuador, 5—SC Chile, 6—Kyushu, 7—N. Sumatra, 8—Alaska, 9—N. Chile, 10—N. Costa Rica, 11—Aleutians, 12—N. Hikurangi, 13—Mariana, 14—Tonga-Kermadec, 15—Kamchatka, 16—Izu, and 17—NE Japan.

A key difference between these results and previous work is that we predict diapiric rise for sediment layers with wet quartz rheology that are as thin as 100 m, whereas Currie *et al.* (ref. 29) concluded that buoyant 'wet quartz' layers less than 1-km thick would not form diapirs unless the magnitude of the density contrast was  $> 200 \text{ kg m}^{-3}$ . Thus, because the sediment layer thickness is < 1 km for all but one of the subduction zones in Fig. 5a, Currie *et al.*<sup>29</sup> would not predict instabilities for any of these unless the density contrast was consistently significantly greater than 200 kg m<sup>-3</sup>. Such high density contrasts are inconsistent with our calculated density contrasts for metasediments at UHP conditions (Fig. 2).

Our calculations predict that in all but four subduction zones with very thin sediment layers, diapirs form within  $\pm 40 \text{ km}$  of

the slab depth below the arc (Fig. 5b). This indicates that partial melts derived from metasedimentary diapirs rising through the hot mantle wedge could easily be incorporated into the arc melting regime. The details of these calculations are sensitive to the slab-top geotherm. Subduction-zone thermal models have evolved considerably over the past 10 years, incorporating effects such as temperature- and stress-dependent rheology, decoupling between the downgoing slab and overriding plate, and variable slab geometry<sup>5-8</sup>. However, many potentially important effects, including thermal advection associated with melts and fluids and thermal/mechanical erosion of the upper plate, have yet to be incorporated into these models. Furthermore, our calculations assume a steady-state thickness of the subducting sediment layer, whereas temporal variations are likely. Thus, although a robust conclusion of our analysis is that diapirs form at temperatures  $<\sim$ 850 °C and near the depth of arc-magma genesis, further details on their formation will require improved subductionzone thermal models.

#### Implications for arc volcanism and mantle wedge dynamics

Based on our scaling analysis (see Methods section), diapirs will form from a sediment layer 250-500 m thick with an along-arc spacing of 4-8 km and diameters of 3-4 km. The diffusion time required to raise the temperature to >1,000 °C at the centre of a sphere with these dimensions is on the order of 10<sup>4</sup> yr assuming that the surrounding mantle is at 1,350 °C. This is more than an order of magnitude faster than the transit time of a similarly sized diapir through the mantle wedge based on analogue experiments<sup>40</sup>, and implies that the sediments would thermally equilibrate and undergo extensive melting before reaching the top of the mantle wedge. However, sediment diapirs of this size would comprise only  $\sim$ 0.1% of the mantle wedge by volume, and thus would have a negligible influence on the time-averaged wedge temperature and the kinematics of mantle flow. Furthermore, given their small size they would be difficult to detect seismically. This contrasts with models of buoyant 'cold plumes' that have been proposed to arise from much thicker layers of hydrated mantle located above or below the downgoing plate<sup>17</sup>, and/or foundering of dense arc lower crust<sup>41</sup>, both of which could significantly alter wedge temperatures and mantle flow patterns. Furthermore, the calculated spacing of sediment diapirs is significantly less than the 30-100 km spacing of most arc volcanoes<sup>32</sup>, indicating that the spacing of sediment diapirs is not the underlying cause of the spacing of volcanic centres in arcs.

The high  $H_2O/Ce$  and  $H_2O/K$  ratios observed in some arcs<sup>11–13</sup> (for example, Tonga, where the rapid convergence rate and great age of the subducting plate combine to yield relatively low slab top temperatures in thermal models) are difficult to explain as the result of extensive melting of metasedimentary diapirs. Instead, it is possible that the subduction component in these arcs is transported from the slab top by means of aqueous fluids or near-solidus, small-degree partial melts, rather than produced by partial melting of diapirs within the mantle wedge. In our study, diapirs are predicted to initiate late, behind the volcanic arc, in the relatively cold Tonga–Kermadec and Mariana–Izu–Bonin arc systems (Fig. 5), where the subducting sediment layer is thin, the recycled sediment signature is muted<sup>2,24</sup>, and recycling efficiencies are relatively low (see Supplementary Information).

Finally, the efficient removal and recycling of sediments into the mantle wedge will influence volatile fluxes into the deep mantle at subduction zones. In the case of H<sub>2</sub>O, only a small fraction of sediment H<sub>2</sub>O (~5%) is subducted to postarc depths<sup>42</sup>, and thus the formation of sediment diapirs will have a relatively minor influence on the total recycling efficiency. However, decarbonation reactions occur at higher temperatures than dehydration reactions, implying that a greater fraction of carbonate rocks may be transported to postarc depths<sup>43–45</sup>. Currently, global estimates of

the slab-derived CO<sub>2</sub> by decarbonation<sup>43</sup> are significantly less than CO<sub>2</sub> drawdown by silicate weathering<sup>46,47</sup>. Thus, sediment diapirs provide an efficient mechanism for recycling these carbonates into the arc melting regime, and may provide a mechanism to balance the global carbon cycle.

## Methods

Sediment densities were calculated using Perple\_X 2007 (ref. 48) and thermodynamic data<sup>49</sup> and solution models<sup>42</sup> appropriate for arc mineral assemblages. We assumed 2 wt% H<sub>2</sub>O for all compositions, which is representative of the amount of H<sub>2</sub>O retained in subducting metasediments at 700 °C and 3 GPa (ref. 42). Based on the calculated density contrast between the average UHP composition and mantle wedge harzburgite<sup>14</sup> at pressures <4 GPa (Fig. 3b) we assume a value of  $-200 \text{ kg m}^{-3}$  for all instability calculations.

The rate of instability growth is sensitive to the viscosity contrast between the buoyant layer and overlying half-space. To determine the viscosity ratio we calculated viscosities for wet quartz<sup>37</sup> and wet olivine<sup>36</sup> ( $C_{OH} = 2,500 \text{ H/10}^6 \text{ Si}$ ) over a range of temperatures and strain rates. For strain rates of  $10^{-16}-10^{-17} \text{ s}^{-1}$  (typical of the slab–wedge interface in numerical models of wedge corner flow with non-Newtonian rheology<sup>5,8</sup>), wet olivine is 2–3 orders of magnitude more viscous than wet quartz at 600–800 °C (the approximate temperature range over which diapirs form). Before 2001, geodynamic studies of density instability assumed that mobile, buoyant crustal layers were more viscous than the adjacent mantle half space, which was then assumed to be inviscid. By contrast, in the scenarios considered here metasediment is much weaker than the mantle. Thus, instability times were calculated based on non-Newtonian growth rates for a 'weak' buoyant layer (layer: half-space viscosity ratio = 1:100) and an assumed background strain rate<sup>35</sup>.

Instability growth rates are sensitive to the length-scale over which viscosity decreases in the overlying half-space<sup>31,34</sup>. For wet olivine, a slab-top temperature of 700 °C, and a vertical temperature gradient of 20–40 °C km<sup>-1</sup> across the slab-wedge interface<sup>6</sup>, the viscous decay length is ~0.5–1.5 km (ref. 34). The instability growth rates were then calculated based on the ratio of the viscous decay length to the sediment layer thickness<sup>35</sup>. Finally, the spacing of the instabilities was determined from the wavenumber of the fastest growth rate.

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#### Author contributions

M.D.B., P.B.K. and G.H. performed the instability calculations. B.R.H., P.B.K. and H-J.M. compiled the UHP metapelite database. P.B.K. compiled the shale and greywacke database and produced the geochemical figures. M.D.B. took the lead in preparing the manuscript with significant input from all authors.

#### Additional information

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