Diapirs as the source of the sediment signature in arc lavas

- 2 Mark D. Behn¹, Peter B. Kelemen², Greg Hirth³, Bradley R. Hacker⁴, and Hans-Joachim Massonne⁵
- ⁴ Department of Geology and Geophysics, Woods Hole Oceanographic Institution, Woods Hole, MA, USA
- 5 Lamont Doherty Earth Observatory, Columbia University, Palisades, NY, USA
- 6 ³Department of Geological Sciences, Brown University, Providence, RI, USA
- 7 Department of Earth Science, University of California, Santa Barbara, California, USA
- 8 ⁵Institut für Mineralogie und Kristallchemie, Universität Stuttgart, Stuttgart, Germany

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Many arc lavas show evidence for the involvement of subducted sediment in the melting process. There is debate whether this "sediment melt" signature forms at relatively low temperature near the fluid-saturated solidus or at higher temperature beyond the breakdown of trace-element-rich accessory minerals. We present new geochemical data from high- to ultrahigh-pressure rocks that underwent subduction and show no significant depletion of key trace elements in the sediment melt component until peak metamorphic temperatures exceeded ~1050°C from 2.7 to 5 GPa. These temperatures are higher than for the top of the subducting plate at similar pressures based on thermal models. To address this discrepancy, we use instability calculations for a non-Newtonian buoyant layer in a viscous half-space to show that, in typical subduction zones, solid-state sediment diapirs initiate at temperatures between 500–850°C. Based on these calculations, we propose that the sediment melt component in arc magmas is produced by high degrees of dehydration melting in buoyant diapirs of metasediment that detach from the slab and rise into the hot mantle wedge. Efficient recycling of sediments into the wedge by this mechanism will alter volatile fluxes into the deep mantle compared to

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Ba, 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 fluids¹.

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 lavas²⁻⁴. Subduction zone thermal models that incorporate temperature- and stress-

estimates based solely on devolatilization of the slab.

33 dependent viscosity⁵⁻⁸ produce slab-top temperatures above the fluid-saturated sediment

^{*}Corresponding Author, Dept. of Geology and Geophysics, Woods Hole Oceanographic Institution, 360 Woods Hole Road MS #22, Woods Hole, MA 02543, email: mbehn@whoi.edu, phone: 508-289-3637, fax: 508-457-2187.

solidus (>600–700°C at \leq 3 GPa; refs ^{9,10}), and H₂O/Ce and H₂O/K ratios in melt 34 inclusions from arc lavas are consistent with fluid fluxed melting of sediments at 750° to 35 more than 950°C beneath several global subduction systems 11-13. Alternatively, it has 36 37 been suggested that subducting sediment detaches as solid-state diapirs, and melts at higher temperatures as it ascends through the mantle wedge¹⁴⁻¹⁷. 38 39 The key to distinguishing between these models is determining the conditions under 40 which sediment melting occurs in subduction zones. A major challenge in evaluating 41 these conditions is the difficulty in sampling the residues of sediment melts. Most studies 42 of rocks exhumed from subduction zones focus on either (1) basaltic compositions 43 analogous to subducted oceanic crust or (2) unusual metasediments that attained high 44 temperatures at relatively low peak pressures, different from even the hottest slab-top 45 geotherms. Mafic ultrahigh-pressure (UHP) rocks subducted to pressure-temperature 46 conditions above the fluid-saturated basalt solidus show MORB-like concentrations in fluid-immobile elements¹⁸, such as Th and La. This suggests that either melting was 47 48 suppressed in these rocks due to low H₂O activity or that minor phases retained Th, La, 49 and other light REE in the solid residue. Similarly, metasediments from Santa Catalina Island¹⁹ show only minor depletion of Th, La, Pb, and Sr. However, these samples 50 51 attained peak pressure-temperature conditions of 600°C and 1–1.2 GPa, a higher 52 temperature at a given pressure than any steady state subduction geotherm, yet below the 53 fluid-saturated sediment solidus. In another example, high pressure and ultrahigh-54 pressure metasediments from the western Alps that reached peak temperatures ≤ 630°C 55 along a plausible subduction geotherm show little sign of depletion of fluid-immobile elements compared to average shale²⁰. 56 57 Characterizing the conditions of sediment melting 58 To characterize the residues of sediment melting, we analyzed and compiled other 59 data on high- to ultrahigh-pressure rocks (peak pressures mostly between 2.7 and 5 GPa; 60 hereafter loosely termed UHP) associated with subduction systems to assess whether 61 these rocks lost Th, Pb, and other fluid-immobile trace elements. In order to isolate 62 pelitic sediment protoliths from igneous compositions we considered only peraluminous 63 UHP compositions (see Supplemental Information for data sources and method for 64 identifying UHP metapelites). Comparing these data to a compilation of peraluminous

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65 shale compositions representative of the incoming sediment at subduction zones (see 66 Supplemental Information for data sources), we find that UHP metapelites with peak 67 temperatures of 700°–1050°C have compositions that lie within the compositional range 68 of shale (Fig. 1a). This indicates that the UHP rocks are indeed representative of 69 peraluminous subducting, pelitic sediments. However, many UHP metapelites that 70 attained temperatures >1050°C show depletions in highly incompatible elements. 71 including Th, Sr, Pb, and Nd, as expected for the residues of melting that could result in 72 extensive recycling of the "sediment component" from subducting metasediments to arc 73 magmas (**Fig. 1** and **Supplementary Figures**). Even among our samples that reached > 74 1050°C, some are not depleted, suggesting that yet higher degrees of melting and/or more 75 efficient melt extraction are required to fully exhaust the minor phases that host key trace 76 elements in the solid residue. 77 Furthermore, we observe phengite within garnet cores in many of our samples at 78 temperatures > 1000°C, indicating that phengite remained stable up to peak metamorphic 79 conditions—as observed experimentally by Hermann and Green (ref ²¹) who report residual phengite at 1000°C at 4 and 4.5 GPa, and as in Schmidt et al. (ref 11) who infer 80 81 that fluid-undersaturated, dehydration melting of phengite occurs from 950°C at 3 GPa to 1150°C at 5 GPa (their Figure 11). Some fluid-saturated²² experiments show lower 82 83 temperatures for phengite breakdown. However, our results indicate that dehydration 84 melting is a better approximation to the conditions in natural UHP metamorphism. 85 Our results are consistent with experimental studies of metasediments and metabasalts 12,23,24, which show little depletion in key trace elements during dehydration 86 87 melting at 600°-1000°C and 2-5 GPa (**Fig. 1b-e**). The lack of depletion is likely due to 88 the stability of minor phases (e.g., phengite, allanite, apatite, monazite, and zircon), 89 which are stable above the fluid-saturated solidus at UHP conditions and retain these key elements²⁴⁻²⁶. 90 91 Overall, the compiled UHP data imply that recycling of elements such as Th, La, Nd 92 and Pb from metasediments is inefficient if slab-top temperatures are < 1000°C in the 93 region of arc magma genesis. Although the parameters involved in the mass balance are 94 uncertain, subducting fluxes of Th, U, K, La and Nd, combined with primitive magmatic concentrations of these elements² and estimates for magmatic fluxes in arcs, generally 95

yield recycling rates of ~100% (see Supplemental Information for these calculations). Thus, the apparent efficiency of recycling together with UHP metapelite compositions suggests that the sediment-melt signature in arcs is generated at >1000°C — significantly above the fluid-saturated sediment solidus. Temperatures greater than 1000°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 >1000°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 1200–1300°C and 4 to 5 GPa (i.e., mantle wedge rather than

Time-scale for the formation of sediment diapirs

subduction zone conditions)²⁷.

Numerical modeling studies have shown the potential for subducting sediments to rise buoyantly into the mantle wedge ^{17,28,29}. However, these studies focused on systems in which the sediments are entrained in buoyant, hydrated mantle diapirs ^{17,28}, or on very thick (> 2 km) sediment layers ²⁹ not representative of the thickness of sediment subducting at modern arcs. Moreover, no studies to date present 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 time-scale and depth at which a sediment layer may go unstable even in the absence of other sources of buoyancy (e.g., 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 ; Supplemental 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 geotherms⁸ the density contrast between the overlying mantle and the average UHP metapelite is as much as -200 kg/m^3 for all pressures up to 6 GPa (**Fig. 3**).

127 The time-scale over which instabilities grow in a buoyant sediment layer is related to 128 the relative viscosities of the sediment and overlying mantle, the viscous decay length in the mantle, and the buoyancy and thickness of the sediment layer³¹⁻³⁵. We calculated 129 instability times assuming a wet olivine rheology for the mantle wedge³⁶ that is 100× 130 131 more viscous than the underlying sediment layer (Fig. 4, see Methods section). This viscosity contrast is appropriate for wet quartz³⁷ at temperatures between 600°–800°C. 132 For a 500 m-thick sediment layer with a density contrast of -200 kg/m³ relative to the 133 134 overlying mantle, the time-scale for instability formation is < 1 Myr and < 10 kyr at 135 temperatures of 700°C and 800°C, respectively (**Fig. 4a** and grey region in **Fig. 4b**). 136 Moreover, any sediment layer thicker than ~100 m will become unstable on time-scales shorter than 1 Myr at temperatures $\leq 1000^{\circ}$ C (**Fig. 4a**). 137 138 The conditions for diapir formation in a given subduction zone can be determined by 139 integrating the instability time along the slab-top pressure-temperature-time (P-T-t) path 140 until the amplitude of the instability exceeds the initial thickness of the sediment layer. 141 To illustrate this approach, the P-T conditions at which diapirs form in Cascadia and Izu— 142 Bonin were calculated assuming a sediment thickness of 500 m (stars on the black & red 143 curves in **Fig. 4b**). In both cases, diapirs form between ~675°-750°C and 2.2-3.0 GPa, 144 with slightly greater formation depth in Izu–Bonin due to the colder incoming slab. Based on slab-top geotherms⁸ and the estimated thickness of the downgoing sediment 145 layer^{38,39}, we explored the conditions of diapir formation for 17 different subduction 146 147 systems (**Fig. 5**). For all subduction systems, diapirs form between 500° and 850°C (**Fig.** 148 5a), with higher temperatures corresponding to thinner sediment layers because instability growth rates scale positively with layer thickness 31,34,35. These temperatures 149 150 are near the fluid-saturated sediment solidus, but significantly below the ~1050°C 151 required to deplete metasediments in Th, Sr, Pb, and Nd (Fig. 1). Thus, we conclude that 152 diapirs of buoyant metasediment detach from the downgoing slab and rise buoyantly into 153 the mantle wedge where they are heated to temperatures > 1000-1100°C. 154 A key difference between these results and previous work is that we predict diapiric 155 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 156 would not form diapirs unless the magnitude of the density contrast was $> 200 \text{ kg/m}^3$. 157

zones in Figure 5A, Currie et al. (ref ²⁹) would not predict instabilities for any of these 159 160 unless the density contrast was consistently significantly greater than 200 kg/m³. 161 Moreover, such high-density contrasts are inconsistent with our calculated density 162 contrasts for metasediments at UHP conditions (Figure 2). 163 Our calculations predict that in all but four subduction zones with very thin sediment 164 layers, diapirs are predicted to form within ± 40 km of the slab depth below the arc (**Fig.** 165 **5b**). This indicates that partial melts derived from metasedimentary diapirs rising 166 through the hot mantle wedge could easily be incorporated into the arc melting regime. 167 The details of these calculations are sensitive to the slab-top geotherm. Subduction-zone 168 thermal models have evolved considerably over the last 10 years, incorporating effects 169 such as temperature- and stress-dependent rheology, decoupling between the downgoing slab and overriding plate, and variable slab geometry⁵⁻⁸. However, many potentially 170 171 important effects including thermal advection associated with melts and fluids and 172 thermal/mechanical erosion of the upper plate have yet to be incorporated into these 173 models. Furthermore, our calculations assume steady-state thickness of the subducting 174 sediment layer, whereas temporal variations are likely. Thus, although a robust conclusion of our analysis is that diapirs form at temperatures < ~850°C and near the 175 176 depth of arc-magma genesis, further details on their formation will require improved 177 subduction-zone thermal models. 178 **Implications for arc volcanism and mantle wedge dynamics** 179 Based on our scaling analysis (see Methods section), diapirs will form from a 180 sediment layer 250–500 m thick with an along-arc spacing of 4–8 km and diameters of 3– 181 4 km. The diffusion time required to raise the temperature to ≥1000°C at the center of a sphere with these dimensions is on the order of 10^4 yr assuming that the surrounding 182 183 mantle is at 1350°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 experiments⁴⁰, 184 185 and implies that the sediments would thermally equilibrate and undergo extensive

Thus, because the sediment layer thickness is < 1 km for all but one of the subduction

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melting before reaching the top of the mantle wedge. However, sediment diapirs of this

size would comprise only ~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 plate¹⁷, and/or foundering of dense arc lower crust⁴¹, both of which could significantly alter wedge temperatures and mantle flow patterns. Further, the calculated spacing of sediment diapirs is significantly less than the 30–100 km spacing of most arc volcanoes³², indicating that the spacing of sediment diapirs is not the underlying cause of the spacing of volcanic centers in arcs.

The high H₂O/Ce and H₂O/K ratios observed in some arcs¹¹⁻¹³ (e.g., 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 via 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^{2,24}, and recycling efficiencies are relatively low (see Supplemental 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_2O , only a small fraction of sediment H_2O (~5%) is subducted to postarc depths⁴², 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 depths⁴³⁻⁴⁵. Currently, global estimates of the slab-derived CO_2 by decarbonation⁴³ are significantly less than CO_2 drawdown by silicate weathering^{46,47}. 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.

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226	Author Contributions
227	MB, PK, & GH performed the instability calculations, BH, PK & HM compiled the UHP
228	metapelite database, PK compiled the shale and greywacke database and produced the
229	geochemical figures, MB took the lead in preparing the manuscript with significant input
230	from all authors.
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233	Methods
234	Sediment densities were calculated using Perple_X 2007 (ref. 48) and thermodynamic
235	data ⁴⁹ and solution models ⁴² appropriate for arc mineral assemblages. We assumed 2
236	wt% H_2O for all compositions, which is representative of the amount of H_2O retained in
237	subducting metasediments at 700°C and 3 GPa (ref. ⁴²). Based on the calculated density
238	contrast between the average UHP composition and mantle wedge harzburgite ¹⁴ at
239	pressures < 4 GPa (Fig. 3b) we assume a value of -200 kg/m^3 for all instability
240	calculations.
241	The rate of instability growth is sensitive to the viscosity contrast between the
242	buoyant layer and overlying half-space. To determine the viscosity ratio we calculated
243	viscosities for wet quartz ³⁷ and wet olivine ³⁶ ($C_{OH} = 2500 \text{ H}/10^6 \text{Si}$) over a range of
244	temperatures and strain-rates. For strain rates of 10^{-16} – 10^{-17} s ⁻¹ (typical of the slab–wedge
245	interface in numerical models of wedge corner flow with non-Newtonian rheology ^{5,8}),
246	wet olivine is 2-3 order of magnitude more viscous than wet quartz at 600-800°C (the
247	approximate temperature range over which diapirs form). Prior to 2001, geodynamic
248	studies of density instability assumed that mobile, buoyant crustal layers were more
249	viscous than the adjacent mantle half space, which was then assumed to be inviscid. By
250	contrast, in the scenarios considered here metasediment is much weaker than the mantle.
251	Thus, instability times were calculated based on non-Newtonian growth rates for a
252	"weak" buoyant layer (layer : half-space viscosity ratio = 1:100) and an assumed
253	background strain rate ³⁵ .
254	Instability growth rates are sensitive to the length-scale over which viscosity
255	decreases in the overlying half-space ^{31,34} . For wet olivine, a slab-top temperature of
256	700°C, and a vertical temperature gradient of 20-40°C/km across the slab-wedge
257	interface ⁶ , the viscous decay length is ~0.5–1.5 km (ref. ³⁴). The instability growth rates
258	were then calculated based on the ratio of the viscous decay length to the sediment layer
259	thickness ³⁵ . Finally, the spacing of the instabilities was determined from the
260	wavenumber of the fastest growth rate.
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263	Figure Captions
264	Figure 1: Comparison of pelitic UHP metasediment compositions to average shale,
265	greywacke and loess compositions. (a) Trace element diagram for pelitic UHP
266	metasediments. 1σ envelope for average shale, greywacke, and loess is shown as blue
267	field with black outlines. (b-e) Trace element concentrations in pelitic UHP
268	metasediments as a function of peak metamorphic temperature compared to our average
269	peraluminous shale and greywacke composition (black circle at arbitrary temperature of
270	600°C), our compiled peraluminous shale, greywacke, and loess compositions (light grey
271	squares and rectangle), and 1σ for our average shale and greywacke composition (dark
272	grey rectangle). Shown for comparison are compositions of residues of partial melting of
273	pelitic red clay ²³ . (See Supplementary Information for data sources.)
274	Figure 2: Density of subducted sediments at UHP conditions. Density calculated at
275	700°C and (a) 1 GPa and (b) 3 GPa as a function of SiO ₂ content for compiled UHP
276	metasedimentary compositions (blue squares), our estimates of average shale and
277	greywacke, average loess, and the average of prior shale compositional averages (red
278	circles), and subducting sediment delineated by lithology (yellow diamonds ³⁰) and
279	location (green diamonds ³⁰). Average continental and arc lower crust ⁵⁰ are shown with
280	black and grey stars, respectively. Black and grey horizontal lines show densities
281	calculated for harzburgite ¹⁴ and pyrolite ³⁵ . Relative to the mantle wedge almost all
282	sediment compositions are buoyant at 700°C and pressures < 3 GPa.
283	Figure 3: Density of the average UHP metasediment along typical subduction zone
284	geotherms. (a) Density contrast between the average UHP metasedimentary
285	composition and mantle wedge harzburgite ¹⁴ as a function of temperature and pressure.
286	Thick red and black lines show slab-top geotherms for Cascadia and Izu-Bonin,
287	respectively ⁸ . (b) Density contrast as a function of pressure along the Cascadia (red) and
288	Izu-Bonin (black) slab-top geotherms. Note that the density contrast is \leq -200 kg/m 3 for
289	all pressures < 6 GPa.
290	Figure 4: Calculated time-scale for the initiation of a sediment diapir. (a) Instability
291	time versus temperature and sediment layer thickness for a density contrast of -200
292	kg/m ³ , background strain-rate of 10 ⁻¹⁶ s ⁻¹ , and initial perturbation amplitude 33% of the

293	layer thickness. For typical subduction rates, diapirs form between 2–4 GPa if the
294	instability time is ≤ 1 Myr (thick black contour). (b) Location of diapir formation (stars)
295	on slab-top geotherms for Cascadia (red) and Izu-Bonin (black). Grey region shows
296	instability times for background strain-rates ranging from 10^{-14} and $10^{-18}~{\rm s}^{-1}$. Diapirs are
297	predicted to form at 685°C and ~2.3 GPa in Cascadia and 730°C and 3.2 GPa in Izu-
298	Bonin.
299	Figure 5: Summary of conditions for sediment diapir formation in global
300	subduction zones. (a) Diapir initiation temperature versus sediment layer thickness ³⁰ ,
301	and (b) diapir initiation depth versus subarc slab depth (as compiled in ref. 8) calculated
302	for 17 slab-top geotherms ⁸ . Subducting sediment layer thicknesses are corrected for
303	compaction to a density of 2800 kg/m ³ . Numbers (in order of increasing slab thermal
304	parameter) correspond to subduction zones: 1 – Cascadia, 2 – Nankai, 3 – Mexico, 4 –
305	Colombia-Ecuador, 5 – SC Chile, 6 – Kyushu, 7 – N. Sumatra, 8 – Alaska, 9 – N. Chile,
306	10 – N. Costa Rica, 11 – Aleutians, 12 – N. Hikurangi, 13 – Mariana, 14 – Tonga-
307	Kermadec, 15 – Kamchatka, 16 – Izu, and 17 – NE Japan.
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