Data report: Pb isotope compositions of sediment from the eastern flank of Juan de Fuca Ridge[1]

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Chapter contents

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Abstract

During Integrated Ocean Drilling Program Expedition 301 we collected sediment samples from the eastern flank (Holes U1301C and U1301D) of Juan de Fuca Ridge. During this expedition, the sediments were squeezed to collect pore water samples, which were analyzed for chemical compositions. The results of Pb isotope analyses on sediment squeezecake samples are presented in this report. The samples have Pb isotopic compositions similar to surface sediments from nearby regions but demonstrate wider variation in $^{206}Pb/^{204}Pb$. In Pb-Pb isotope space, ratios form a trend subparallel to altered turbidites from the northern Juan de Fuca Ridge. The low $^{206}Pb/^{204}Pb$ end of the trend extends toward the seawater field.

Introduction

During Ocean Drilling Program (ODP) Leg 168, 10 sites were drilled across the eastern flank of the Juan de Fuca Ridge to examine conditions of fluid-rock interaction in three distinct hydrothermal regimes extending over a ~120 km linear transect perpendicular to the spreading ridge (Davis, Fisher, Firth, et al., 1997). The main aim of Integrated Ocean Drilling Program (IODP) Expedition 301 to the eastern flank of the Juan de Fuca Ridge was to evaluate the formation-scale hydrogeologic properties within oceanic crust, determine how fluid pathways are distributed within an active hydrothermal system, and elucidate relations between fluid circulation, crustal stratigraphy, alteration, microbiology, and seismic properties (see the **"Expedition 301 summary"** chapter).

In this report, we present data for Pb isotopic analysis of representative sediment samples, which are squeezed residues that remained after taking pore water, collected from IODP Site U1301 (Fig. **F1**). Study of lead isotopes is important for understanding seafloor hydrothermal processes, including leaching of oceanic crust during high-temperature alteration, and the source of metals in hydrothermal deposits. The relatively impermeable sediment cover over the spreading center traps hydrothermal fluids and allows chemical interaction between fluids and sediments. For instance, sediment-hosted massive sulfide deposits, currently forming at the sediment-filled Middle Valley segment of the northern Juan de Fuca Ridge, have Pb isotopic compositions intermediate

between Juan de Fuca basalts and Middle Valley turbiditic sediments (Cousens et al., 2002), indicating that hydrothermal fluids have interacted with the sediment pile.

Samples and analytical procedures

Sediment samples were obtained using the advanced piston corer and maintained under a nitrogen atmosphere during the squeezing process. After squeezing, sediment samples were freeze-dried and then crushed into a fine powder and homogenized with an agate mortar and pestle.

Reagent-grade deionized water (18 MΩ) was obtained using a Milli-Q purification system. High-purity acids (HF, HNO_3, HCl, and HBr) used in this study were Tamapure-AA-100 grade (Tama Chemicals Co., Ltd., Tokyo) and were used as received or diluted with reagent water.

Approximately 100 mg samples of finely powdered sediment were dissolved using a mixture of HF and HNO_3. Dissolution was performed on a hotplate (~100°C; overnight) in closed 7 mL polytetrafluoroethylene (PTFE) Savilex beakers with 1.5 mL of 14 M HNO_3 and 3 mL of 29 M HF. The sample solution was evaporated to dryness on a hotplate. The residue was then treated overnight with 1 mL of 6 M HCl at ~90°C. After evaporation to dryness, the residue was dissolved in ~1 mL of 0.6 M HBr.

Pb was separated from the sample solution by conventional HBr/HCl techniques using PTFE microcolumns (Manhes et al., 1978), Muromac AG 1-X8 anion-exchange resin (100–200 mesh; ~0.06 mL resin bed; Muromachi Technos Co., Ltd., Tokyo), and two column passes for sample purification.

Fractionation correction for Pb isotope measurement was done using the ^{207}Pb–^{204}Pb double spike technique. The ^{207}Pb–^{204}Pb double spike (^{207}Pb and ^{204}Pb = ~5 ppm; $^{207}Pb/^{204}Pb$ = 0.992001) of the University of the Ryukyus was made by mixing of single ^{207}Pb (^{207}Pb = 92.81%) and ^{204}Pb (^{204}Pb = 99.73%) spike solutions. Single spikes were prepared by dissolution of metal-form Pb purchased from Oak Ridge National Laboratory (USA), with 0.3 M HNO_3. Calibration of double spike isotopic composition was performed relative to the National Institute of Standards (NIST) standard reference material (SRM)-982 Pb, assuming $^{208}Pb/^{206}Pb$ is 1.00016 for this standard (Todt et al., 1996; Thirlwall, 2000). Calculation for mass bias correction followed the method in the appendix of Galer (1999), which assumes a linear mass fractionation law.

Following Woodhead et al. (1995), the chemically separated Pb sample was dissolved in dilute HNO_3 and split between the natural and mixed runs immediately before loading onto a previously outgassed flat Re filament. The double spike solution (~1 μL) was added directly to the "mix" filament, and 3 μL of emitter solution, similar to Gerstenberger and Haase (1997), was added to both natural and mixed filaments, with mixing encouraged by repeatedly sucking the mixture back into the loading pipette.

Pb isotope composition was determined using a Finnigan MAT262 mass spectrometer at the University of the Ryukyus. Data acquisition was performed on ^{208}Pb ion beam intensities between 1.5×10^{-11} and 2.5×10^{-11} A. During the experiment period, NIST SRM-981 Pb yielded average values of 16.940 ± 0.004 (2 σ; $N = 4$) for $^{206}Pb/^{204}Pb$, 15.497 ± 0.004 for $^{207}Pb/^{204}Pb$, and 36.718 ± 0.011 for $^{208}Pb/^{204}Pb$. These values compare well with the preferred values obtained by double spike (e.g., Thirlwall, 2000) and triple spike (Galer and Abouchami, 1998) techniques. Total procedural Pb blanks were generally <30 pg. Because blank contribution is significantly low (<<0.003%) relative to Pb amount (several μg), blank effects on measured isotopic ratios are negligible.

Results

Lead isotope data on sediment samples were 18.913–19.172 for $^{206}Pb/^{204}Pb$, 15.632–15.675 for $^{207}Pb/^{204}Pb$, and 38.743–39.092 for $^{208}Pb/^{204}Pb$ (Table T1). Three samples from Sections 301-U1301C-4H-5, 11H-1, and 301-U1301D-5H-1 are described as slightly more radiogenic ($^{206}Pb/^{204}Pb$ > 19.1) than other samples ($^{206}Pb/^{204}Pb$ < 19.1) (Fig. F2).

The Pb isotopic variation with depth profile of sediment cores is shown in Figure F3. With the exception of the three radiogenic samples ($^{206}Pb/^{204}Pb$ > 19.1), the sediment samples display a slight decline in $^{206}Pb/^{204}Pb$ with increasing depth, whereas $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ are almost unchanged.

The field defined by these sediment samples departs from the Juan de Fuca and Gorda Ridge basalt field (Fig. F2). The sediment data plot close to the field of surface sediments from the northern Juan de Fuca Ridge (Cousens et al., 2002) but demonstrate a wider range of variation. They are distinct from altered turbidites from the northern Juan de Fuca Ridge, although the trends defined by sediment samples are subparallel to the turbidites field. An interesting point is that the trends defined by these sediment samples extend at the lower $^{206}Pb/^{204}Pb$ end toward the Pacific seawater field represented by Mn crust (von Blanckenburg et al., 1996). Conversely, massive

sulfides from the northern Juan de Fuca Ridge (Cousens et al., 2002) plot between the basalts field and sediments with steeper trend; Pb isotopic composition of the most radiogenic massive sulfides overlap our sediment samples with the lowest $^{206}Pb/^{204}Pb$.

Acknowledgments

References

Cousens, B.L., 1996. Depleted and enriched upper mantle sources for basaltic rocks from diverse tectonic environments in the northeast Pacific Ocean: the generation of oceanic alkaline versus tholeiitic basalts. *In* Basu, A.R., and Hart, S.R. (Eds.), *Earth Processes: Reading the Isotopic Code.* Geophys. Monogr., 95:207–231.

Cousens, B.L., Blenkinsop, J., and Franklin, J.M., 2002. Lead isotope systematics of sulfide minerals in the Middle Valley hydrothermal system, northern Juan de Fuca Ridge. *Geochem., Geophys., Geosyst.,* 3(5):1029. **doi:10.1029/2001GC000257**

Davis, E.E., Fisher, A.T., Firth, J.V., et al., 1997. *Proc. ODP, Init. Repts.,* 168: College Station, TX (Ocean Drilling Program). **doi:10.2973/odp.proc.ir.168.1997**

Fouquet, Y., and Marcoux, E., 1995. Lead isotope systematics in Pacific hydrothermal sulfide deposits. *J. Geophys. Res.,* 100:6025–6040. **doi:10.1029/94JB02646**

Galer, S.J.G., 1999. Optimal double and triple spiking for high precision lead isotopic measurement. *Chem. Geol.,* 157(3–4):255–274. **doi:10.1016/S0009-2541(98)00203-4**

Galer, S.J.G., and Abouchami, W., 1998. Practical application of lead triple spiking for correction of instrumental mass discrimination. *Mineral. Mag.,* 62A(1):491–492. **doi:10.1180/minmag.1998.62A.1.260**

Gerstenberger, H., and Haase, G., 1997. A highly effective emitter substance for mass spectrometric Pb isotope ratio determinations. *Chem. Geol.,* 136(3–4):309–312. **doi:10.1016/S0009-2541(96)00033-2**

Hart, S.R., 1984. A large-scale isotope anomaly in the Southern Hemisphere mantle. *Nature (London, U. K.),* 309(5971):753–757. **doi:10.1038/309753a0**

Hegner, E., and Tatsumoto, M., 1987. Pb, Sr, and Nd isotopes in basalts and sulphides from the Juan de Fuca Ridge. *J. Geophys. Res.,* 92(B11):11,380–11,386. **doi:10.1029/JB092iB11p11380**

Manhes, G., Minster, J.F., and Allègre, C.J., 1978. Comparative uranium-thorium-lead and rubidium-strontium study of the Saint Sèverin amphoterite: consequences for early solar system chronology. *Earth Planet. Sci. Lett.,* 39(1):14–24. **doi:10.1016/0012-821X(78)90137-1**

Thirlwall, M.F., 2000. Inter-laboratory and other errors in Pb isotope analyses investigated using a ^{207}Pb–^{204}Pb double spike. *Chem. Geol.,* 163(1–4):299–322. **doi:10.1016/S0009-2541(99)00135-7**

Todt, W., Cliff, R.A., Hanser, A., and Hoffmann, A.W., 1996. Evaluation of a ^{202}Pb–^{205}Pb double spike for high-precision lead isotopic analysis. *In* Basu, A.R., and Hart, S.R. (Eds.), *Earth Processes: Reading the Isotopic Code.* Geophys. Monogr., 95:429–437.

von Blanckenburg, F., O'Nions, R.K., and Hein, J.R., 1996. Distribution and sources of pre-anthropogenic lead isotopes in deep ocean water from Fe–Mn crusts. *Geochim. Cosmochim. Acta,* 60(24):4957–4963. **doi:10.1016/S0016-7037(96)00310-9**

Woodhead, J.D., Volker, F., and McCulloch, M.T., 1995. Routine lead isotope determinations using a lead-207–lead-204 double spike: a long-term assessment of analytical precision and accuracy. *Analyst,* 120(1):35–39. **doi:10.1039/an9952000035**

Figure F1. Eastern flank of Juan de Fuca Ridge showing location of ODP Leg 168 Sites 1026 and 1027 and IODP Site U1301. Samples reported in this study are from Site U1301. FR = first ridge, SR = second ridge, DR = deep ridge.
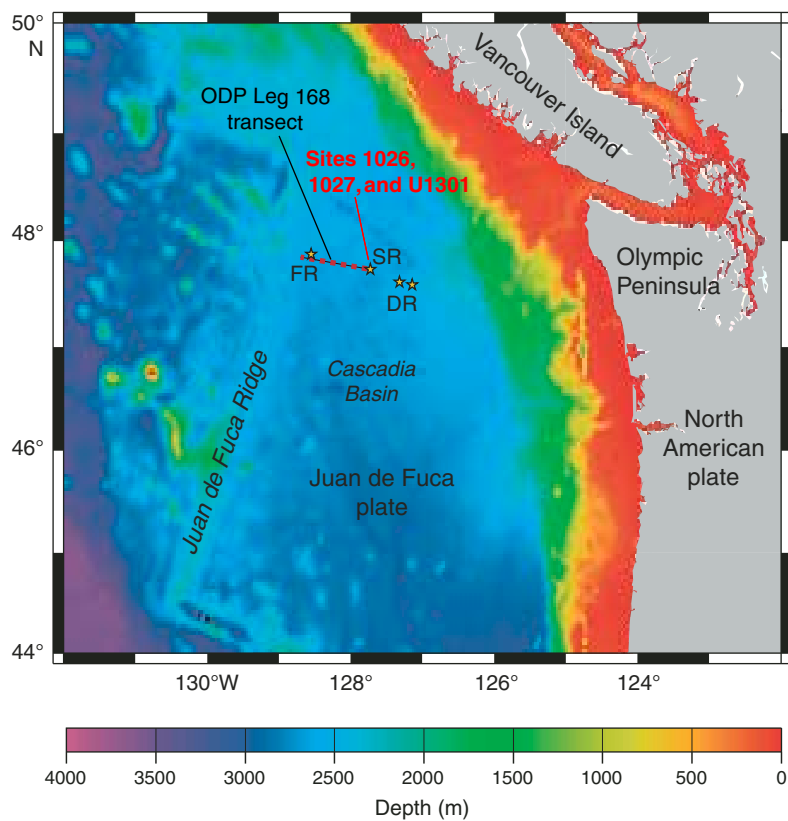
Figure F2. Pb-Pb isotope plots. Solid circles = sediment samples from this study, open circles = massive sulfides from the Middle Valley of the north end of the Juan de Fuca Ridge (Cousens et al., 2002). Juan de Fuca/Gorda Ridge basalts field data compiled by Cousens (1996). Juan de Fuca Ridge bare-rock sulfide data from Fouquet and Marcoux (1995) and Hegner and Tatsumoto (1987). Turbidite and surface sediment data from Cousens et al. (2002). Pacific seawater (represented by Mn crust) data from von Blanckenburg et al. (1996). NHRL = Northern Hemisphere Reference Line (Hart, 1984).
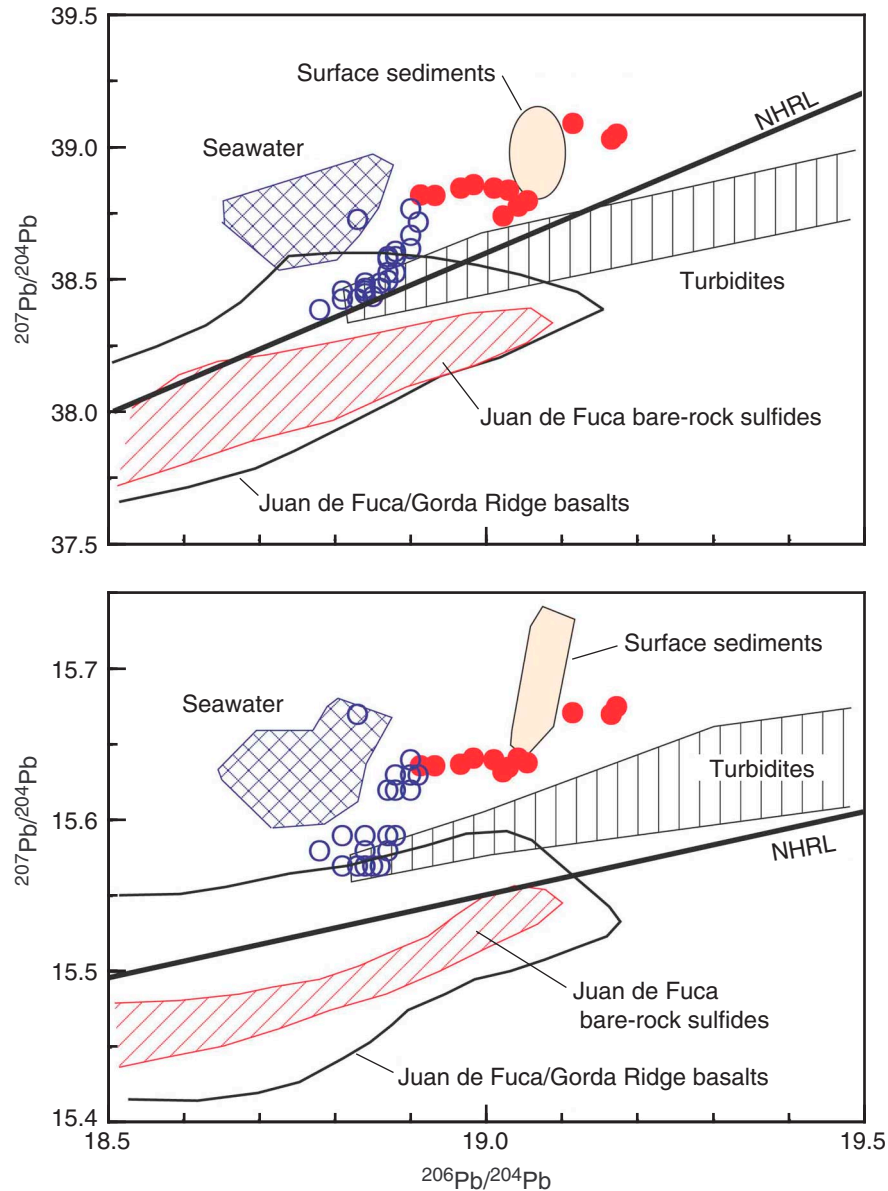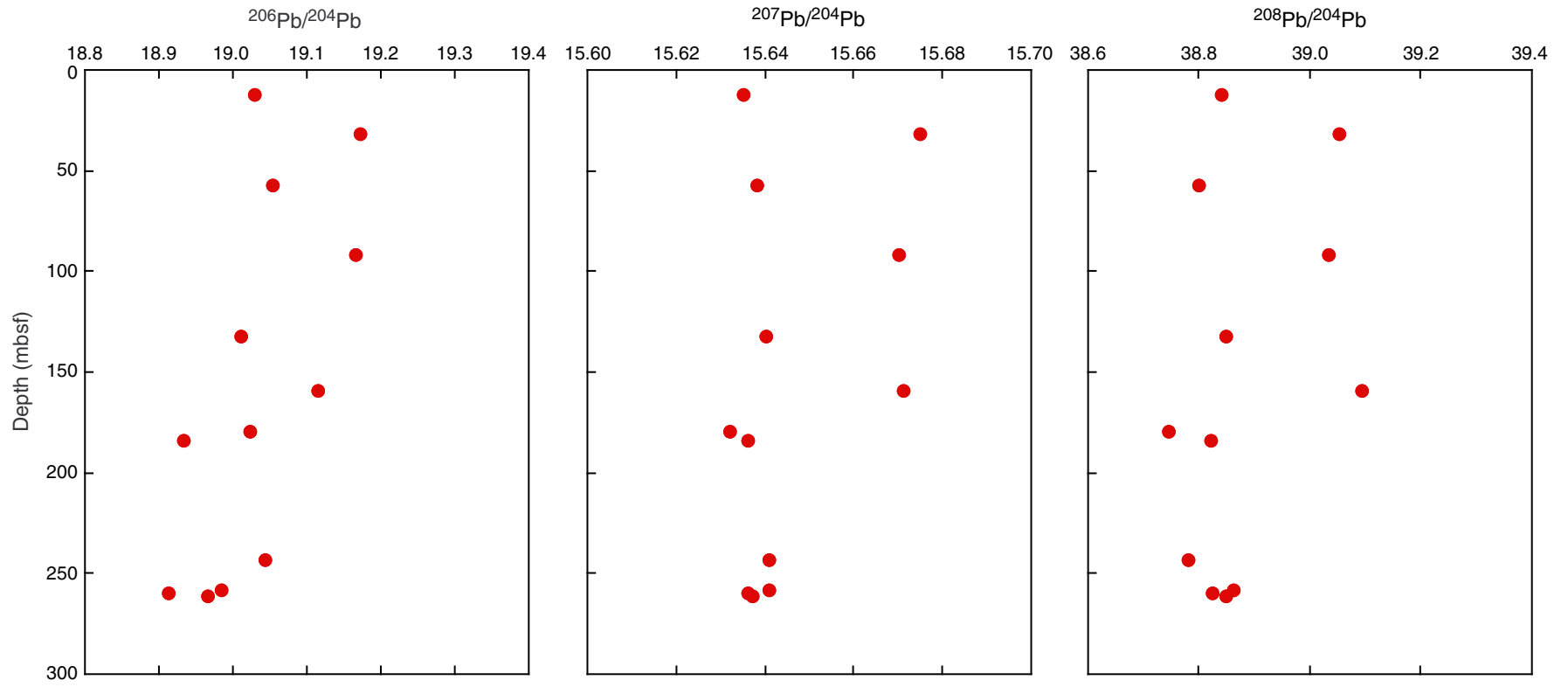
Figure F3. Pb isotopic variation vs. depth.

Table T1. Pb isotopic compositions, Site U1301. (**See table note.**)

Core, section, interval (cm)	Depth (mbsf)	$^{206}Pb/^{204}Pb$ (±2 σ)	$^{207}Pb/^{204}Pb$ (±2 σ)	$^{208}Pb/^{204}Pb$ (±2 σ)
301-U1301C-				
2H-5, 130–150	12.4	19.029 ± 2	15.635 ± 1	38.840 ± 4
4H-5, 130–150	31.3	19.172 ± 2	15.675 ± 2	39.051 ± 4
7H-3, 110–150	56.9	19.054 ± 2	15.638 ± 2	38.800 ± 4
11H-1, 110–150	91.7	19.165 ± 2	15.670 ± 2	39.033 ± 4
15H-1, 110–150	179.2	19.022 ± 2	15.632 ± 2	38.743 ± 4
15H-4, 110–150	183.7	18.932 ± 2	15.636 ± 1	38.820 ± 3
17H-5, 110–150	242.9	19.042 ± 2	15.641 ± 2	38.780 ± 4
19H-2, 110–150	258.4	18.983 ± 1	15.641 ± 1	38.861 ± 3
19H-3, 110–150	259.9	18.913 ± 2	15.636 ± 2	38.822 ± 4
19H-4, 110–150	261.4	18.966 ± 2	15.637 ± 1	38.848 ± 3
301-U1301D-				
2H-2, 110–150	131.8	19.010 ± 1	15.640 ± 1	38.848 ± 3
5H-1, 110–150	159.1	19.114 ± 2	15.671 ± 1	39.092 ± 3

Note: Internal precision (2 σ) of each run based on internal errors propagated through the double spike mass bias correction and corresponds to the last digit.