sea level and to an arbitrary reference composition. Each normalized value is given by

$$(R_{\mathrm{m}} - R_0) \frac{\mathrm{Cl_m}}{\mathrm{Cl_r}} \left[\frac{\mathrm{K_2O_r}\ (\psi_{\mathrm{K}}) + \mathrm{CaO_r}\ (\psi_{\mathrm{Ca}})}{\mathrm{K_2O_m}\ (\psi_{\mathrm{K}} + \mathrm{CaO_m}\ (\psi_{\mathrm{Ca}})} \right. \left. \frac{+\ [\mathrm{Cl}]_{\mathrm{r}}\ (\psi_{\mathrm{n}})_{\mathrm{r}}}{+\ [\mathrm{Cl}]_{\mathrm{m}}\ (\psi_{\mathrm{n}})_{\mathrm{m}}} \right] \frac{1}{ELD} \qquad (3)$$

where the subscripts m and r indicate a measured or reference value, respectively, and where the concentration of Cl ([Cl]) is in parts per million (ppm). Samples C84-41 and C84-40 (Table 1) were normalized from 2.0 m depth to land surface using densities of 2.60 and 2.35 g $\mathrm{cm^{-3}}$, respectively (on the basis of specific gravity measurements), and a half-attenuation length of 132 g $\mathrm{cm^{-2}}$ (*9*). The reference composition was 2.35 percent $\mathrm{K_2O}$, 5.00 percent CaO, and 165 ppm chlorine. The reference production rate by neutron activation of $^{35}\mathrm{Cl}$ (ψ_{n}) was 81 atoms of $^{36}\mathrm{Cl}$ per kilogram per year per parts per million of chlorine. R_0 was calculated for each sample as described (*7*).

In Fig. 1 the normalized $^{36}\mathrm{Cl/Cl}$ ratios are compared with the theoretical buildup curve for the reference rock calculated from Eq. 2. The samples show a consistent buildup of $^{36}\mathrm{Cl}$ with time. Although the number of samples analyzed is small, they do demonstrate that buildup of cosmogenic radionuclides can be measured in rocks at the earth's surface. With further confirmation, we expect that $^{36}\mathrm{Cl}$ buildup can be used to date surface exposure times. If so, this would open the possibility of determining the age of a wide variety of materials that were not previously subject to direct dating. The ability to date directly the exposure time of objects such as archeological artifacts, young volcanic rocks, glacial moraines, geomorphic surfaces, and soils would constitute a powerful tool for numerous disciplines within the earth sciences.

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Hydrothermal Germanium Over the Southern East Pacific Rise

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Germanium enrichment in the oceanic water column above the southern axis of the East Pacific Rise results from hydrothermal solutions emanating from hot springs along the rise crest. This plume signature provides a new oceanic tracer of reactions between seawater and sea floor basalts during hydrothermal alteration. In contrast to the sharp plumes of $^3\mathrm{He}$ and manganese, the germanium plume is broad and diffuse, suggesting the existence of pervasive venting of low-temperature solutions off the ridge axis.

THE PREDICTION OF SEA FLOOR HYdrothermal activity at spreading centers (*1*) and the subsequent discovery of hot springs along mid-ocean ridges (*2, 3*) led to successful explanations linking the process to the existence of metalliferous sediments, "missing" conductive heat flow near ridge crests, ophiolite complexes, seafloor basalt alteration (*4, 5*), and mid-water hydrothermal plumes (*6–9*). These phenomena are all associated with convection of seawater to great depths through fissures in the oceanic crust where reactions between seawater and basalts at high temperatures and pressures produce highly altered metabasalts and hydrothermal solutions (*4*). The chemistry of these solutions expelled back to the oceans through vents along the mid-ocean ridge crest is dramatically different from that of the original seawater, providing a mechanism for transfer of elements between the earth's crust and oceans of potentially enormous significance (*10*).

We describe enrichments in dissolved inorganic germanium of up to 25 percent above that expected for the deep Pacific Ocean. These enrichments are due to emanations of germanium-enriched hydrothermal solutions (plumes) along the ridge axis of the southern East Pacific Rise crest. This discovery provides the possibility of linking the inventory of hydrothermally derived germanium in the oceans with the systematics of its extraction from oceanic basalts to yield an additional constraint on the circulation of seawater through the global mid-ocean ridge–hydrothermal system, the extent of basalt alteration, and the consequent fluxes of elements into or out of the oceans. There is considerable uncertainty over the magnitude of these fluxes (*4, 11*). Germanium may prove to be a useful tracer of hydrothermal processes because of its geochemistry: it is a trace element closely allied with silicon (*12*) and thus provides a powerful new tool for investigating crustal processes (*13*).

Vertical profiles of the germanium concentration anomaly (ΔGe) over the East Pacific Rise axis (Fig. 1) outline plumes of water that are enriched above local seawater in inorganic germanium (*14*). The magnitude of the largest anomaly (25 p*M*) represents an enrichment of about 25 percent above local background germanium concentrations. The germanium plume is located at mid-depth (~2500 m) and corresponds roughly to the depths of the hydrothermally tagged $^3\mathrm{He}$, manganese, and iron plumes (*7–9*). In contrast to the distinctly sharp plumes observed for manganese and $^3\mathrm{He}$, however, the ΔGe signature over the East Pacific Rise crest is broad and diffuse.

The locations of the stations between 12°S and 20°S coincide with a hydrother-

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mally active section of the ridge axis, as evidenced by fast spreading rates (*15*), rapidly accumulating metalliferous sediments (*16, 17*), and large ^{3}He anomalies (*7*). The anomaly appears to be a maximum at 20°S and to decrease nonuniformly northward along the ridge axis, parallel to decreasing gradients in spreading rates (*15*) and accumulation of metalliferous sediments (*16*). Hence, the gradient in ΔGe roughly parallels the gradient of hydrothermal activity along the crest of the East Pacific Rise.

The departure of these East Pacific Rise data from the general background Ge/Si ratio recycled within the ocean, although less than 25 percent, is statistically significant (Fig. 2). We made every effort to exclude the possibility that these data are systematically higher because of an undetected analytical artifact or a hidden blank (*18*). More data will be required to confirm the absolute magnitude and spatial extent of the true anomaly.

There is ample evidence that solutions resulting from hydrothermal processes are enriched in germanium compared to seawater and river water concentrations and compared to typical Ge/Si ratios observed in the oceans, rivers, and crustal rocks. Deep-sea water exhibits germanium concentrations of 50 to 100 p*M* and a constant Ge/Si of 0.7×10^{-6}, whereas uncontaminated rivers show germanium concentrations of 50 to 300 p*M* and Ge/Si ratios of about 0.3 to 1.0×10^{-6} (*13*). Concentrations of germanium in crustal rocks are typically 1 to 2 ppm, with Ge/Si ratios of 1 to 2.6×10^{-6}. In contrast, vent samples from the Galápagos Rift (*10*) display a Ge/Si ratio of 16×10^{-6}, with germanium concentrations of up to 16 n*M* (*13*). Germanium concentrations in the black smokers at 21°N on the East Pacific Rise crest (*3, 19*) are about 150 n*M*, enriched more than 1500 times the concentrations in Pacific deep waters. The Ge/Si ratios in these East Pacific Rise vents are about 9×10^{-6} (*20*). Germanium concentrations in Icelandic geothermal waters (*21*) range from 30 to 400 n*M*, with Ge/Si ratios of 100×10^{-6} to 300×10^{-6}. Comparable germanium enrichment has been observed in hot springs from Japan, Bulgaria, and the Soviet Union (*22*); in geothermal well samples from Mauna Loa; and in hot springs from the Azores and the Andes (*23*). Thus hydrothermal solutions, whether terrestrial or marine or in granitic or basaltic terrain, are ubiquitously enriched in germanium compared to solutions resulting from low-temperature alteration.

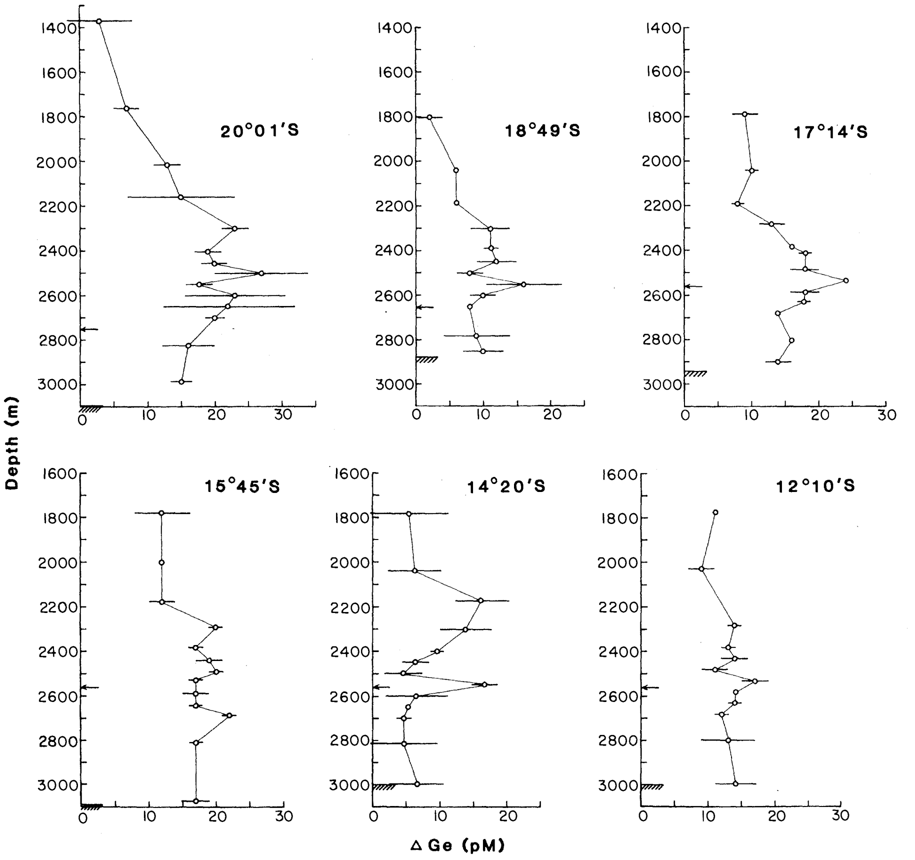

Fig. 1. Vertical profiles of the germanium concentration anomaly (ΔGe) along the southern axis of the East Pacific Rise crest. Samples were collected for ^{3}He, manganese, and germanium during cruise TT 154 of the R.V. *Thompson* (*14*). Station locations are 20 01.2'S, 113 42.3'W (TT 154-017); 18 48.9'S, 113 28.5'W (TT 154-016); 17 13.8'S, 113 14.8'W (TT 154-015); 15 44.8'S, 112 49.5'W (TT 154-014); 14 20.4'S, 112 38.2'W (TT 154-013); and 12 10.0'S, 110 51.3'W (TT 154-012). ΔGe is the excess germanium concentration above that expected from the oceanic Ge/Si relation and the observed silicon concentration: $\Delta\text{Ge} = \text{Ge}_\text{A} - \text{Ge}_\text{B}$ (p*M*), where ΔGe is the germanium contribution from the hydrothermal plume (anomaly), Ge_B is the oceanic recycled Ge concentration (p*M*), $\text{Ge}_\text{B} = 0.71 \times 10^{-6}\,(\text{Si}_\text{B}) + 3.8$ (*13*), Si_B is the silicon concentration (in micromoles per kilogram); corrected for a very small hydrothermal silicon contribution given by $\text{Si}_\text{B} = \text{Si}_\text{A} - 0.07\,\Delta^3\text{He}$ (*10*). Si_A and Ge_A are the observed silicon (in micromoles per kilogram) and inorganic germanium (p*M*) concentrations. Silica data are from Murray (*14*). Horizontal bars and data points denote either the range of duplicate or standard deviation of triplicate determinations. Symbols without bars have a standard deviation less than the size of the data point. Arrows on the left axis mark the depth of the top of the tallest seamount at the same latitude observed by sonar (*14*). Hatched symbols on the left axis denote the depth of the sea floor directly underneath each hydrocast.

Since germanium is widely dispersed in silicate minerals in a uniform ratio to silicon (camouflage, *12*), its enrichment in hydrothermal solutions cannot be simply the result of congruent dissolution unless there are germanium-enriched minerals in the hydrothermal mineralization zone that do not survive to be found in oceanic basalts (*24*). Initially we suspected that sea-floor hydrothermal solutions might display higher Ge/Si ratios than continental rivers because of a ratio in oceanic basalts two to three times that in continental granites (*13, 25*). We now propose that the large enrichments we observe are due to dissolution followed by extreme germanium depletion in secondary silicate minerals forming within hydrothermal vent plumbing. There are three lines of evidence for this. First, Ge/Si ratios of Icelandic geothermal waters are orders of magnitude higher than the ratio in rocks being altered (*21*). Second, silica scale and sinter precipitating in the Hawaiian and Azorean thermal systems are 700 to 300 times as depleted in Ge/Si as their companion solutions (*23*). Arnórsson (*21*) observed similar depletions in Icelandic silica sinter. Third, germanium may be carried in hydrothermal solutions as soluble Ge II, perhaps as GeS (*21, 26*), rather than as Ge(OH)_4 (Ge IV). Germanium in hydrothermal systems is chalcophilic, not lithophilic: it is not sequestered into high-temperature secondary silicate minerals but is instead associated with hydrothermal sulfides (*21, 27*). Thus precipitation of secondary hydrothermal silicate minerals, depleted in germanium, can produce elevated Ge/Si ratios in the resulting solutions.

The source of the germanium anomaly over the East Pacific Rise crest cannot be simply due to the conversion of seawater

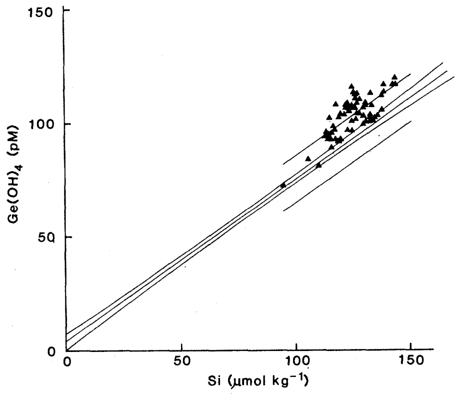

Fig. 2. Germanium and silicon values (▲) from cruise TT 154 are plotted on the regression line for all oceanic data (*13*). The background oceanic data are described by the linear regression $Ge = 0.71 \times Si + 3.8$; $r^2 = 0.998$ ($n = 122$). The 99 percent confidence band about this regression is given by the curved lines. A confidence interval of 2 standard deviations about the oceanic Ge data (±10 p*M*) is given by the parallel straight lines between 100 and 150 for silicon. The mean value of ΔGe in the East Pacific Rise data between 2200 m and the sea floor (Fig. 1) is 14 p*M* and represents a real enrichment in germanium compared to the oceanic trend ($P < 0.0001$). Note that ΔGe is approximately equal to the vertical difference between data points (▲) and the Ge/Si regression line.

organogermanium [monomethylgermanium (MMGe) and dimethylgermanium (DMGe)] to inorganic germanium by thermal decomposition upon passage through the ridge crest. The methylated germanium compounds show constant concentrations in the ocean of about 330 p*M* of MMGe and 120 p*M* of DMGe, and thus are present in seawater in concentrations about five times those of inorganic germanium at the depth of the ridge crest (*28*). A ΔGe anomaly of 25 p*M* requires the mixing of about one part hydrothermal water (Ge = 150 n*M*) with 5000 parts local seawater (Ge = 100 p*M*). Hence, if all seawater methylgermanium is converted to inorganic germanium, it can contribute only 0.3 percent of the anomaly, clearly insignificant compared to that extracted from basalts.

The existence of this germanium anomaly requires that the Ge/Si ratio entering seawater from sea floor vents be greater than the oceanic recycled ratio. Since the $Ge/^3He$ ratio in these East Pacific Rise plumes (*29*) is similar to that observed directly in vent exhalations (*30*), germanium apparently mixes conservatively into bottom waters with little removal into scavenging metal oxides (*13*) or precipitating sulfides, somewhat surprising considering Ge's tendency to associate with sphalerite in hydrothermal mineralizations. If germanium and silicon are both nearly conservative after exhalation to the ocean, then based on the $Ge/^3He$ ratio in the plumes and the $^3He/Si$ ratio observed directly in vents, we estimate that the hydrothermally derived Ge/Si atom ratio entering the ocean is about 7×10^{-6} (*31*), nearly ten times as enriched in Ge as both the oceanic recycled ratio (0.7×10^{-6}) (*13*) and the continentally derived river ratio (0.5×10^{-6}) (*23*).

The Ge/Si ratios observed directly in 21°N East Pacific Rise black smokers (9×10^{-6} at 350°C) and in the Galápagos "warm" vents (16×10^{-6} at <20°C) and estimated from the plume signature (7×10^{-6}) are all greatly enriched over the marine tholeiitic basalt ratio (2.6×10^{-6}) (*25*). These data suggest that significant quantities of silicon are partitioned into secondary hydrothermal silicate minerals at the expense of germanium, perhaps progressively so in regimes with extensive mixing between high-temperature end-members and cold ground seawater within the vent plumbing (for example, the Galápagos) (*5*). This interpretation is consistent with notions that most ^3He and manganese (and other elements) are extracted and expelled to seawater through localized high-temperature vents (ridge axis "smokers"), but that significant germanium and heat (plus other elements but little ^3He and manganese) are emitted from pervasive off-axis effluents at lower temperatures to produce a diffuse, cloudlike germanium anomaly such as we observed. If this interpretation is correct, then the hydrothermal source Ge/Si ratio estimated from the plume anomaly is a lower limit because the off-ridge vents likely carry a $Ge/^3He$ ratio higher than that from ridge axis smokers.

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30. $(Ge)_{vents} = 150$ n*M* (*20*); $(^3He)_{vents} = 7.7 \times 10^{-12}$ mole per liter, estimated from ^3He/heat ratio in Galápagos vents extrapolated to 350°C [W. J. Jenkins, J. M. Edmond, J. B. Corliss, *Nature (London)* **272**, 156 (1978)]; therefore $(Ge/^3He)_{vents} = 1.9 \times 10^4$ moles of germanium per mole of ^3He.
31. $(Ge/Si)_{HT\text{-}source} = (Ge/^3He)_{plumes} \times (^3He/Si)_{vents}$; $(^3He/Si)_{vents} = 3.6 \times 10^{-10}$ (mole per mole) (*10*); therefore $(Ge/Si)_{HT\text{-}source} = 7 \times 10^{-6}$ mole of germanium per mole of silicon.
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