Coupled ^{186}Os and ^{187}Os Evidence for Core-Mantle Interaction

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Osmium isotopic analyses of picritic lavas from Hawaii show enrichments in the osmium-186/osmium-188 ratio (^{186}Os/^{188}Os) of 0.008 to 0.018%, relative to a chondritic upper mantle, that are positively correlated with enrichments in ^{187}Os/^{188}Os of 5.4 to 9.0%. The most viable mechanism to produce these coupled ^{186}Os and ^{187}Os enrichments is by addition of 0.5 to 1 weight percent of outer core metal to a portion of the D″ layer and subsequent upwelling of the mixture. These data suggest that some plumes originate at the core-mantle boundary and that Os isotopes may be used to distinguish plumes derived from shallow versus deep mantle sources.

The depth of initiation of hot, upwelling plumes in Earth's mantle is not well defined (*1*, *2*), although plumes likely originate at thermal or chemical boundaries within the mantle or at the core-mantle boundary. The D″ layer (*3*) at the core-mantle boundary is evidently a very chemically active region of the Earth where some level of metal-silicate interaction almost certainly occurs (*4*, *5*). If some plumes originate at the core-mantle boundary, then certain geochemical characteristics of the outer core may be imparted to the rising plume and subsequent plume-derived lavas.

Outer core contributions to a plume can potentially be detected by examination of highly siderophile elements (HSEs) in plume-derived lavas (*6*) because of the extreme contrast in the concentrations of HSEs between the outer core and mantle and the likelihood that the HSEs are fractionated relative to one another in the outer core compared with the generally chondritic relative abundances of these elements in the upper mantle (*7*, *8*). Such fractionations would be characteristic of solid metal–liquid metal equilibrium. Hence, the addition of outer core metal to a plume may impart a unique HSE signature to the plume that cannot be reproduced by processes that occur within the mantle (*7*). However, crystal-liquid fractionation processes that may occur during ascent and emplacement of plumes and derivative magmas may obscure such elemental signals.

Concentration data for three HSEs in iron meteorites, Re, Pt, and Os, show that progressive asteroidal core crystallization produced melts with increasingly suprachondritic Re/Os and Pt/Os ratios (*7*, *9*). If similar partitioning occurred in the Earth's core during crystallization of the inner core, then plumes enriched in outer core material should show coupled enrichments in ^{187}Os/^{188}Os and ^{186}Os/^{188}Os ratios, reflecting the decay of ^{187}Re and ^{190}Pt to ^{187}Os and ^{186}Os, respectively (*10*, *11*). The large differences in the concentrations of HSEs between the outer core and the mantle means that additions of only 0.5 to 1 weight % of outer core metal to a plume source would significantly dominate the Os isotopic composition (*11*). Unlike the relative abundances of HSEs, the isotopic composition of Os is not changed by crystal-liquid fractionation processes.

By this reasoning the suprachondritic ^{187}Os/^{188}Os isotopic compositions of about 0.130 to 0.150 in ocean island basalts (OIBs) relative to the chondritic average ratio of 0.1270 in the upper mantle might, in part, reflect outer core entrainment (*7*, *9*, *12*). However, because the Re/Os ratio of oceanic crust is also suprachondritic, entrainment of 1- to 3-billion-year-old (Ga) oceanic crust may also generate the radiogenic ^{187}Os/^{188}Os ratio observed in plume-derived lavas (*12*–*17*). Hence, ^{187}Os enrichment alone is not a definitive test for core-mantle interaction and corresponding ^{186}Os data are needed (*18*).

We have determined ^{186}Os/^{188}Os ratios for picritic lavas from Hawaii and compared these compositions with Os-Ir alloys and chromitites from presumed upper mantle–derived ultramafic systems (*19*) to determine whether an outer core component is present in the Hawaiian plume source. These Hawaiian lavas are suitable to test for core-mantle interaction because they are variably enriched in ^{187}Os, with γ_{Os} values ranging from +5.2 to +9.1 (*15*–*17*) (Table 1), and because Hawaiian magmas are derived from one of the hottest and longest-lived mantle plumes documented (*20*).

Table 1. Re and Os isotopic data for Os-Ir alloys and chromitites from peridotite massifs and ophiolites worldwide and picrites from Hawaii. Number of unspiked replicates for high-precision ^{186}Os/^{188}Os and ^{187}Os/^{188}Os analyses is listed in parentheses after the sample name. The $2\sigma_M$ for the average of the replicate runs is reported as precision in the sixth place after the value. ^{187}Os/^{188}Os for the spiked run (SP) is reported with run statistics.

Sample	Re (ng/g)	Os (ng/g)	^{186}Os/^{188}Os	^{187}Os/^{188}Os	γ_{Os}	ε_{Nd}
Os-Ir alloys						
W5A (3)			0.1198342 ± 29	0.124268 ± 43	−2.15	
W5B (3)			0.1198356 ± 34	0.124252 ± 23	−2.16	
California (3)			0.1198355 ± 31	0.123329 ± 18	−2.89	
Savage River (3)			0.1198361 ± 15	0.119940 ± 09	−5.56	
Bald Hills (3)			0.1198304 ± 24	0.120041 ± 22	−5.48	
19 Mile Creek (3)			0.1198346 ± 08	0.123824 ± 21	−2.50	
Urals (3)			0.1198306 ± 02	0.119861 ± 03	−5.62	
Chromitites						
HPOS 33 (3)	1.33*	79.28*	0.1198330 ± 37	0.12378 ± 52	−2.54	
HPOS 74 (3)	3.35*	147.2*	0.1198339 ± 33	0.12734 ± 48	+0.27	
HPOS 498 (3)	4.08*	236.1*	0.1198365 ± 38	0.12247 ± 36	−3.56	
Hawaiian picrites						
Mauna Loa						
ML-2-50 (4)	0.433†	0.792†	0.1198425 ± 32	0.133878 ± 35	+5.41	+5.5†
ML-1868.9 SP	0.631	0.717		0.13453 ± 14	+5.93	
ML-1868.9 (3)			0.1198409 ± 12	0.135499 ± 03	+6.69	
Loihi						
LO-02-02 (4)	0.610†	0.863†	0.1198443 ± 28	0.133606 ± 324	+5.20	+5.5†
LO-02-04 (2)	0.481†	1.083†	0.1198433 ± 17	0.134266 ± 352	+5.72	+3.9†
Koolau						
KOO-17A (3)	0.224†	0.465†	0.1198526 ± 34	0.138499 ± 06	+9.05	+0.2†

*From (*32*). Chromitites are from the following ophiolites: HPOS 74, Semail (Oman); HPOS 33 and 498, Al'Ays (Saudi Arabia).

†Concentration data from (*17*).

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Thus, the Hawaiian plume is a manifestation of a long-lived thermal anomaly potentially resulting from upwelling of material from the deep mantle. In addition, the lavas we have studied are picrites with high Os abundances ranging from 0.47 to 1.08 ng/g (*17*) (Table 1). This is important because magmas with such high Os concentrations are unlikely to have been significantly contaminated by Os from the oceanic lithosphere (*14–17*).

The Os-Ir alloys and chromitites we examined have $^{187}Os/^{188}Os$ ratios ranging from 0.11986 to 0.12734 (Table 1). Most of these samples are at the more radiogenic end of this range, consistent with derivation from an upper mantle having Os isotopic compositions within the range of abyssal peridotites and chondrites (0.1220 to 0.1273) (*21*). Several Os-Ir alloys have lower $^{187}Os/^{188}Os$ ratios of 0.1199 to 0.1220, consistent with derivation from a source that lost Re by melt removal 1 to 2 Ga (Table 1). The average $^{186}Os/^{188}Os$ for these 10 samples is 0.1198340 ± 12 ($2\sigma_M$). The $^{186}Os/^{188}Os$ ratio of 0.1198310 ± 60 for the carbonaceous chondrite Allende (*9*) is identical, within uncertainty, to the $^{186}Os/^{188}Os$ ratios for the chromitites and the Os-Ir alloys. These similarities indicate <30% fractionation of the long-term Pt/Os ratio for the upper mantle relative to chondrites, even in materials that experienced ancient melt depletion. Evidently Pt and Os behave compatibly during partial melting of the upper mantle, and Pt is not fractionated from Os in the depleted upper mantle.

In contrast to the samples from the upper mantle, the Hawaiian picrites all have excess ^{186}Os and positive γ_{Os} values (Table 1, Fig. 1). The $^{186}Os/^{188}Os$ and $^{187}Os/^{188}Os$ ratios for the picrites are more radiogenic than our upper mantle average and are consistent with variable long-term coupled Pt/Os and Re/Os enrichment in the plume source of the Hawaiian lavas. The enrichments in ^{186}Os are positively correlated with enrichments in ^{187}Os, forming a linear array that extends from a composition enriched in ^{186}Os and ^{187}Os toward a composition that appears to be enriched in $^{187}Os/^{188}Os$ by about 2% relative to the average chondritic value but with chondritic $^{186}Os/^{188}Os$ (Fig. 1). Uncertainties in the data, however, cannot preclude chondritic values of both $^{187}Os/^{188}Os$ and $^{186}Os/^{188}Os$ in this hypothetical end member. This array requires at least two source components. One component reflects long-term enrichment in both Pt and Re relative to the upper mantle, and the other may have generally chondritic Pt/Os but with a possibly more radiogenic $^{187}Os/^{188}Os$ than the chondritic average.

The covariance of $^{186}Os/^{188}Os$ and $^{187}Os/^{188}Os$ ratios in the picrites is very similar to that observed in iron meteorites (*7*) and may be explained by mixing of outer core material into the plume source. In fact, the most radiogenic sample (Koolau) has nearly the same composition for both $^{186}Os/^{188}Os = 0.119852$ and $\gamma_{Os} = +9.05$ as that calculated for the outer core by the asteroidal analog (*9*, *11*). Although the absolute Os isotopic composition of the outer core is not known because of the large temperature and pressure contrasts between asteroidal cores and the terrestrial core, and the growth rate of the inner core, the relative partitioning between the HSEs is likely not greatly different from the lower pressure systems (*9*, *11*). The outer core component, however, cannot be much more radiogenic than the Koolau picrite (*22*).

Alternatively, the radiogenic Os isotopic compositions of the Hawaiian picrites may reflect an ancient recycled oceanic crustal component in the plume. Because Re abundances in basalts and sediments are up to several hundred times higher than mantle peridotite (*23*), the addition of recycled oceanic crust to a mantle reservoir can lead to the formation of a hybrid mantle with suprachondritic Re/Os and, with time, suprachondritic $^{187}Os/^{188}Os$ (*14–17*). Additions of up to 30% of ancient ocean crust to the sources of some plumes have been advocated to produce the observed $^{187}Os/^{188}Os$ ratios of up to ~0.150 in OIB (*14–16*). For example, addition of 6% of a recycled component consisting of 90% 1.8-Ga oceanic crust plus 10% pelagic sediment to the plume source has been proposed to produce $^{187}Os/^{188}Os$ of 0.138 in some lavas from Hawaii (*16*).

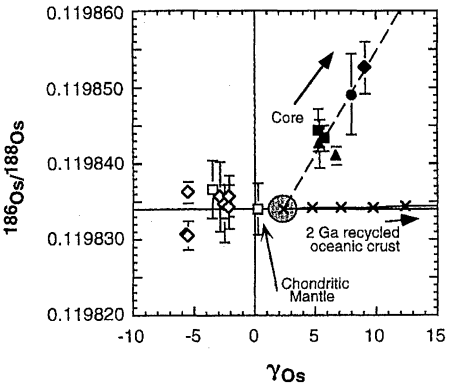

Fig. 1. $^{186}Os/^{188}Os$ versus γ_{Os} for the Noril'sk ore (●) (*9*), Os-Ir alloys (◇), chromitites (□), and the Loihi (■), Mauna Loa (▲), and Koolau (◆) picrites. Error bars for $^{186}Os/^{188}Os$ are 2σ. Error bars for γ_{Os} are smaller than the symbols. A mixing line is shown with 2% increments (×) of 2-Ga recycled ocean crust, consisting of 90% altered oceanic basalt (Os = 0.1 ng/g, $^{187}Os/^{188}Os$ = 5) and 10% pelagic sediment (Os = 0.2 ng/g, $^{187}Os/^{188}Os$ = 0.5) added to a plume source mantle (Os = 3 ng/g, $^{187}Os/^{188}Os$ = 0.130) as described in (*16*). For altered ocean crust, we use a Pt/Os ratio of 19.84 for MORB (Fig. 2), which, if allowed to evolve 2 Ga after separation from a chondritic mantle, will have a present day $^{186}Os/^{188}Os$ = 0.1198810. For pelagic sediment, we use a Pt/Os ratio of 89.33, the same as upper crust in Figure 2 because it is the highest Pt/Os ratio estimated for sediments and will result in the most elevated $^{186}Os/^{188}Os$ of 0.1200641 after separation from a chondritic mantle at 2 Ga. Nevertheless, this mixing line is nearly flat and the Hawaiian-Noril'sk array cannot be reproduced by a reasonable Pt/Os ratio and Pt abundance for recycled ocean crust. Dashed line is the inferred mixing line between the Hawaiian plume source (*16*) and a radiogenic $^{186}Os/^{188}Os$ and $^{187}Os/^{188}Os$ likely to be the outer core.

Although recycling of crustal materials can lead to ^{187}Os enrichments within a mantle reservoir, this could not be the primary process responsible for generating the compositions of Hawaiian lavas because it would not lead to a corresponding enrichment in ^{186}Os of the magnitude observed. Although Pt/Os ratios for basalts and sediments can be quite high (10 to 1000 times the chondritic ratio), the Pt concentrations in these rocks are not enriched over the upper mantle. Upper mantle peridotites contain Pt concentrations of 3 to 10 ng/g (*24*), basalts typically have about 0.2 to 10 ng/g (*25*, *26*), and sediments typically range from about 0.5 to 10 ng/g (*26*). This limited range in Pt contents between mantle materials and possible recycled components, coupled with the fact that Os is a highly compatible element during partial melting of the mantle, means that large increases in the Pt/Os of a mantle domain are not likely to occur by crustal recycling. For example, the most radiogenic samples from Hawaii, with $^{186}Os/^{188}Os$ of 0.119846 to 0.119852, would require a 2-Ga source that is 80 to 95% sediment or basalt (Fig. 2). A source with such large proportions of basalt or sediment would produce siliceous (>50 weight % SiO_2) melts and not the picritic Hawaiian lavas examined here. Primitive Hawaiian magmas have MgO contents of at least 12 to 15 weight % MgO and are in equilibrium with Fo_{88-90} olivine, consistent with melting of a dominantly peridotitic source (*27*). Such large percentages of recycled materials with high Re/Os would also generate more radiogenic $^{187}Os/^{188}Os$ ratios (γ_{Os} = 270 to >3000) (*28*) than are observed. Another problem with the generation of the excess ^{186}Os by crustal recycling is that there is growing evidence that, when slabs dehydrate or melt beneath volcanic arcs, Re, Os, and Pt may be mobilized and concentrated into the water and in chlorine-rich fluids and melts that are transferred from the slab into the mantle wedge (*29*). Thus, subducted oceanic crust may have lower concentrations of Re, Os, and Pt than the relatively pristine materials that have been used to define the modeling parameters (Fig. 2) (*26*, *30*).

We conclude that the coupled ^{186}Os-^{187}Os systematics in the Hawaiian picrites is evidence for low and variable degrees of outer core entrainment in the plume source. The lithophile isotopic characteristics of associated Hawaiian tholeiites require processes other than simple addition of core material to a plume source composed of upper mantle similar to the source of mid-ocean ridge basalts (MORB). Koolau basalts, which show the strongest evidence for core interaction in their Os isotopic compositions, also have Pb (^{206}Pb/^{204}Pb = 17.8), Sr (^{87}Sr/^{86}Sr = 0.7045), and Nd (ε_{Nd} ~ 0) isotopic compositions that are distinct from those of MORB (*31*), and none of these characteristics can be explained by entrainment of core material into the plume. The nature and origin of the Koolau component with respect to these lithophile isotopes is currently under debate (*1*, *16*, *17*) and may consist of either ancient recycled crust (*1*, *16*) or primitive lower mantle (*17*), or it may reflect refertilization of a portion of the mantle by melts derived from mafic reservoirs (*17*). Thus, it may be possible that a limited proportion of the ^{187}Os enrichment in Hawaiian lavas comes from a recycled crustal component, as has been previously suggested for OIB from the Azores (*12*). However, the proportion of Re added to a hybrid source is probably overestimated in the models proposed to account for ^{187}Os enrichments in OIB (*14*–*16*), so the recycled component likely would have had only a minor effect on the Os isotopic composition of the system. This is particularly the case for ^{186}Os/^{188}Os (Fig. 2). Therefore, although the origin of the lithophile isotopic composition of the low ε_{Nd}, low ^{206}Pb/^{204}Pb end member in the Hawaiian lavas can be attributed to one of several processes, none of which reflects outer core additions to the plume, the radiogenic ^{186}Os and ^{187}Os end member in the Hawaiian plume is best explained by 0.5 to 1 weight % addition of lithophile element–poor, HSE-rich outer core metal.

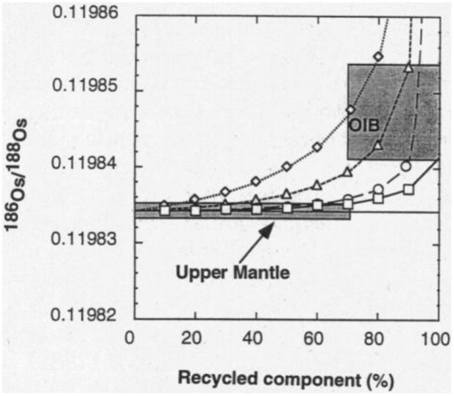

Fig. 2. Mixing relationships for adding 2-Ga ocean crust [sediments and MORB] to a mantle source having ^{186}Os/^{188}Os = 0.1198340 and Os at 3 ng/g. Upper continental crust (△) is calculated taking the Os abundance (0.03 ng/g) and Pt abundance (2.68 ng/g) interpolated from Pd and Ru from (*27*). For reducing sediments (□), Os = 0.244 ng/g, and Pt = 1.16 ng/g; for metalliferous sediments (◇), Os = 0.153 ng/g and Pt = 7.41 ng/g, and for MORB, Os = 0.05 ng/g, and Pt =0.992 ng/g. The Pt and Os abundances for the sediments are averages of data, and the Pt and Os abundances for MORB (○) were chosen from the median of basalts and picrites from (*25*). These components are assumed to have chondritic ^{186}Os/^{188}Os at 2 Ga = 0.1198288 [calculated using a present day ratio of 0.119834, a chondritic ^{190}Pt/^{188}Os = 0.001692 (*9*), and a decay constant of ^{190}Pt = 1.541×10^{-12} a^{-1} (*9*)] and evolve to present day values with the calculated ^{190}Pt/^{188}Os ratios. Symbols on the mixing lines are for 10% increments 2-Ga ocean crust.

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6. The HSEs consist of Re, Au, and the platinum group elements—Os, Ir, Pt, Ru, Rh, and Pd. They are so named because of their strong affinity with iron.
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10. ^{187}Re decays to ^{187}Os by $^{-}\beta$ transition with a decay constant of 1.666×10^{-11} a^{-1} [M. I. Smoliar, R. J. Walker, J. W. Morgan, *Science* **271**, 1099 (1996)]. ^{190}Pt undergoes α decay to ^{186}Os with a decay constant of 1.541×10^{-12} a^{-1} (*9*).
11. Core formation sequestered most of Earth's HSE [H. E. Newsom and K. Sims, *Science* **252**, 926 (1991)]. Consequently, the bulk core likely has chondritic relative abundances of HSE and about a factor of 10^3 higher concentrations than the upper mantle. Metal from primitive H5 ordinary chondrites has 3000 ng of Os per gram and ^{187}Re/^{188}Os ratios near the chondritic average of 0.402 [J.-M. Luck and C. J. Allegre, *Nature* **302**, 130 (1983)]. This is assumed to be the initial composition of the Earth's core (*7*). The upper mantle is estimated to have an Os concentration of 3.3 ng/g [J. W. Morgan, *J. Geophys. Res.* **91**, 12375 (1986)]. Inner core crystallization has likely caused fractionation of the HSE in the outer core relative to the chondritic average and lower concentrations of the HSE in the outer core than in the bulk core. Modeling predicts a decrease of Os from ~3000 to 1040 ng/g in the outer core during 5.5% fractional crystallization, which contrasts with ~3 to 4 ng of Os per gram contained in the upper mantle. Hence, the HSE concentrations in the outer core are likely 300 to 400 times higher than HSE concentrations in the mantle. At 5.5% crystallization of the inner core with initial ratios for ^{187}Re/^{188}Os = 0.4224 and a chondritic ^{190}Pt/^{188}Os = 0.001692 (*7*, *9*), the outer core would have ^{187}Re/^{188}Os = 0.5303 and ^{190}Pt/^{188}Os = 0.00423. If crystallization of the inner core occurred within several hundred million years after core separation, the outer core would evolve to present-day values of ^{187}Os/^{188}Os = ~0.1372 (corresponding to γ_{Os} = +8.0; where γ_{Os} is the percent deviation from the chondrite average of ^{187}Os/^{188}Os = 0.1270 for samples with present day crystallization ages) and ^{186}Os/^{188}Os = 0.119852, using initial chondritic ratios. Recent low-pressure experimental evidence has verified that Pt behaves less compatibly than Os during crystallization of metal from an iron liquid for compositions relevant for the Earth's core, consistent with observations from the iron meteorites (H. V. Lauer Jr. and J. H. Jones, *Lunar Planet. Sci. Conf.* **XXIX**, 1796 (1998). Solid metal–liquid metal partition coefficients for HSEs are not sensitive to pressure, so these low-pressure experiments and the iron meteorite data may accurately reflect partitioning behavior at the higher pressures of Earth's core crystallization (J. H. Jones and D. Walker, *Earth Planet. Sci. Lett.* **105**, 127 (1991).
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18. High-precision measurements of ^{186}Os/^{188}Os from upper mantle Os-Ir alloys and a platinum-group element-rich ore from the Noril'sk area intrusions, a portion of the plume that generated the Siberian Flood Basalt Province, showed that the ^{187}Os-enriched Siberian plume with γ_{Os} = +8.5, was about 0.012% enriched in ^{186}Os relative to the chondritic upper mantle (*9*). The coupled enrichments are consistent with the addition of outer core metal with elevated Re/Os and Pt/Os to the Siberian plume (*7*, *9*, *11*).
19. ^{190}Pt is a minor isotope of platinum (0.0124 atomic %). This combined with a small decay constant (*10*) limits the variation of ^{186}Os/^{188}Os in nature and, for mantle samples considered in this investigation, differences can be detected only by measurements that are 2 orders of magnitude more precise than those for ^{187}Os/^{188}Os. For unspiked high-precision Os isotope analyses, the following procedures were used. The amount of Os necessary to sufficiently perform precise measurements is 30 to 40 ng. For picrites and ultramafic rocks with ~0.3 to 4 ng of Os per gram, 10 to 100 g of powder was fused by a standard nickel sulfide fire assay procedure [E. L. Hoffman, A. J. Naldrett, J. C. Van Loon, R. G. V. Hancock, A. Mason, *Anal. Chem.* **102**, 157 (1978)]. The rock powder was mixed with a flux of sodium tetraborate, ultrapure Ni powder, and sublimed sulfur powder in a ceramic crucible and heated to 1100°C to form a homogenous silicate melt with immiscible NiS melt. Os in the rock powder was partitioned into the NiS beads that form in the bottom of the crucible. The melt was

quenched and the NiS beads were extracted and dissolved in 12 N HCl. The HCl solution was filtered through a cellulose membrane that traps insoluble Os sulfide. The cellulose membranes were then dissolved and standard Os distillation techniques or, later in the study, solvent extraction [A. S. Cohen and F. G. Water, *Anal. Chem.* **332**, 269 (1996)] were used to purify the Os, followed by microdistillation to further purify the Os. Blanks are 1 pg per gram of sample fused and have ^{186}Os/^{188}Os = 0.1199 ± 0.0002, and ^{187}Os/^{188}Os = 0.125 ± 0.005. Corrections to the sample ratios using these values are well within the uncertainties quoted in Table 1. Total procedural yields were 75 to 95 weight %. For Re and Os concentration determinations, 1- to 2-g samples of powder were spiked and dissolved in aqua regia in Carius tubes [S. B. Shirey and R. J. Walker, *Anal. Chem.* **67**, 2136 (1995)] to obtain sample spike equilibration followed by distillation or solvent extraction. Negative thermal ionization mass spectrometer procedures for unspiked high precision Os and spiked Re and Os measurements are presented elsewhere [(7) and references therein]. For high-precision Os analyses, we used a multicollector dynamic Faraday cup mode. Beam voltages ranged from 50 to 150 mV on ^{186}Os and ^{187}Os for at least 100 ratios to achieve the desired internal precision of 40 to 80 parts per million (2σ) on ^{186}Os/^{188}Os. Tungsten trioxide, a possible interference at masses 232 and 234 (^{184}Os/^{188}Os and ^{186}Os/^{188}Os, respectively), was monitored via mass 232 (232/236 ratio) and was not observed under the run conditions. Multiple analyses of a Johnson and Matthey standard solution measured in this way gave external precisions of ^{186}Os/^{188}Os = 0.1198473 ± 40 and ^{187}Os/^{188}Os = 0.1137830 ± 50 (n = 38) during the course of the study.

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22. The ^{186}Os/^{188}Os of the proposed outer core component could not be much greater than the calculated value of 0.119852, assuming ≤1 weight % core addition to a plume. Crystallizing the inner core with proportionally greater or lesser partition coefficients for Re, Pt, and Os could lead to greater or lesser enrichment in ^{186}Os and ^{187}Os. However, enrichments would necessarily also be reflected in lower Os concentrations in the outer core (7). Although the outer core could be more radiogenic, if the lower mantle has approximately the same Os concentration as the upper mantle, the effects would be diluted by the mantle component.
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Distribution of Rock, Metals, and Ices in Callisto

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Radio Doppler data from a single encounter (C3) of the Galileo spacecraft with Callisto, the outermost Galilean moon of Jupiter, indicated that Callisto was probably undifferentiated. Now, similar data from a second encounter (C9) corroborate this conclusion, but more accurate data from a third encounter (C10) indicate that the rock and ice within Callisto have partially, but not completely, separated. Callisto may be differentiated into a rock-metal core less than 25 percent of Callisto's radius, an outer layer of clean ice less than 350 km thick, and a middle layer of mixed rock and ice. Models in which ice and rock are mixed all the way to the center of Callisto are also consistent with the data.

During the primary Galileo Orbiter mission, three close flybys (C3, C9, C10) of Callisto provided data on this moon's gravitational field (*1*). Using radio Doppler data generated by the Deep Space Network (DSN) at three 70-meter stations located at Goldstone, California (DSS14), near Madrid, Spain (DSS63), and near Canberra, Australia (DSS43), and using nonlinear weighted least squares (2), we determined the second-degree coefficients in the standard spherical harmonic expansion of the gravitational potential, V (3).

In principle, the rotational and tidal distortions of Jupiter's Galilean satellites are separately excited and separately measurable. Thus it is possible to infer the degree of differentiation of each satellite in two independent ways (*4*). For a satellite such as Callisto, in synchronous rotation with its orbital period, equilibrium theory predicts that the gravitational coefficient $J_2 \equiv -C_{20}$ is exactly 10/3 of C_{22} [see (3) for the definition of C_{20} and C_{22}]. Any significant deviation from this relationship indicates that the assumption of hydrostatic equilibrium is not appropriate. However, because the three flybys did not provide a global coverage of the satellite's gravitational field (Table 1), an independent determination of J_2 and C_{22} was impossible. We applied the 10/3 constraint for all fits to the data.

The flyby geometry was more favorable for C3 than for C9 or C10, because along-track and cross-track components of the Doppler shift could be detected. For C9, the spacecraft passed directly behind Callisto (an Earth occultation), and could not communicate with stations on Earth during the occultation. As a result, there is a gap of about 11 min in the C9 data at closest approach. The altitude of the spacecraft above Callisto at occultation ingress was 1473 km and at egress was 1103 km. For C10 the spacecraft passed directly in front of Callisto, and radio Doppler data were obtained before, during, and after closest approach (Fig. 1). However, any gravitational perturbations for C9 and C10 were detected by the Callisto-centered trajectory bending only, while for C3 the bending and the velocity perturbation along the orbital path were detected.

The sensitivity of the radio Doppler data to Callisto's gravity depends on whether or not the data were coherent with atomic frequency standards at the DSN stations. This coherency was achieved only when the spacecraft radio system was locked to a signal from a DSN station by means of its S-band transponder. Otherwise, the data were referenced to the spacecraft's crystal oscillator with its inherently poor frequency stability, unknown frequency bias, and unknown frequency drift, in comparison to atomic frequency standards. In fitting the C3 and C9 noncoherent data we included the bias and drift as parameters in the model. However, because the bias and drift were negligible for the atomic-referenced coherent data, they were not included in the C10 model. Even though the geometry was more favorable for C3, the coherent data made C10 a better candidate for a reliable determination of Callisto's gravitational field.

The least-squares solution for J_2 and C_{22} from the C3, C9, and C10 data, analyzed independently (Table 1), depends on the assumption that all other harmonics in the

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