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Mercury abundances and isotopic compositions in the Murchison (CM) and Allende (CV) carbonaceous chondrites

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Abstract—The abundance and isotopic composition of Hg was determined in bulk samples of both the Murchison (CM) and Allende (CV) carbonaceous chondrites using single- and multi-collector inductively coupled plasma mass spectrometry (ICP-MS). The bulk abundances of Hg are 294 ± 15 ng/g in Murchison and 30.0 ± 1.5 ng/g in Allende. These values are within the range of previous measurements of bulk Hg abundances by neutron activation analysis (NAA). Prior studies suggested that both meteorites contain isotopically anomalous Hg, with $\delta^{196/202}\text{Hg}$ values for the anomalous, thermal-release components from bulk samples ranging from -260 ‰ to $+440$ ‰ in Murchison and from -620 ‰ to $+540$ ‰ in Allende (Jovanovic and Reed, 1976a; 1976b; Kumar and Goel, 1992). Our multi-collector ICP-MS measurements suggest that the relative abundances of all seven stable Hg isotopes in both meteorites are identical to terrestrial values within 0.2 to 0.5 ‰.

On-line thermal-release experiments were performed by coupling a programmable oven with the single-collector ICP-MS. Powdered aliquots of each meteorite were linearly heated from room temperature to 900°C over twenty-five minutes under an Ar atmosphere to measure the isotopic composition of Hg released from the meteorites as a function of temperature. In separate experiments, the release profiles of S and Se were determined simultaneously with Hg to constrain the Hg distribution within the meteorites and to evaluate the possibility of Se interferences in previous NAA studies. The Hg-release patterns differ between Allende and Murchison. The Hg-release profile for Allende contains two distinct peaks, at 225° and 343°C , whereas the profile for Murchison has only one peak, at 344°C . No isotopically anomalous Hg was detected in the thermal-release experiments at a precision level of 5 to 30 ‰, depending on the isotope ratio. In both meteorites the Hg peak at $\sim 340^\circ\text{C}$ correlates with a peak in the S-release profile. This correlation suggests that Hg is associated with S-bearing phases and, thus, that HgS is a major Hg-bearing phase in both meteorites. The Hg peak at 225°C for Allende is similar to release patterns of physically adsorbed Hg on silicate and metal grains. Prior studies suggested that the isotopic anomalies reported from NAA resulted from interference between ^{203}Hg and ^{75}Se . However, the amount of Se released from both meteorites, relative to Hg, is insufficient to produce all of the observed anomalies. Copyright © 2001 Elsevier Science Ltd

1. INTRODUCTION

Hg is an intriguing element to study in meteorites because it: (a) is highly volatile, (b) has seven stable isotopes with a relatively large mass range (196–204 amu), (c) forms bonds that have a high degree of covalent character, and (d) exists in more than one oxidation state. These factors suggest that it may undergo kinetic isotopic fractionation in natural systems (Kuznetsov and Obolenskii, 1980; O'Neil, 1986). Hg is predicted to condense at or below 525 K in the solar nebula by chemisorption on metal grain surfaces (Lauretta et al., 1999). HgS occurs at metal-sulfide interfaces in an unusual Allende xenolith, consistent with low-temperature condensation from a S-rich nebular gas (Kurat et al., 1989). Based on observations of HgS in the Mighei CM chondrite, Hg can be concentrated by low-temperature aqueous alteration on chondrite parent bodies (Lauretta et al., 1999). Thus, Hg isotopes may record signatures of low-temperature events in the solar nebula and on meteorite parent bodies.

The cosmochemical behavior of Hg has been the subject of study for over 40 y. Its abundance and isotopic composition has

been measured in chondrites, achondrites, iron meteorites, Martian meteorites, and lunar samples (see compilation by Lauretta et al., 1999). Published measurements of both the Hg abundances and the $^{196}\text{Hg}/^{202}\text{Hg}$ isotope ratios scatter by more than 3 orders of magnitude, even for samples of the same meteorite. As a result, many questions remain regarding its distribution in extraterrestrial materials.

Every report of isotopically anomalous Hg is based on neutron activation analysis (NAA). While this technique is capable of measuring very low concentrations of Hg (ng/g), it can only detect two of the seven stable isotopes (^{196}Hg and ^{202}Hg). Thus, only anomalies in the $^{196}\text{Hg}/^{202}\text{Hg}$ ratio have been reported. Even though the reported anomalies are controversial, they cannot be summarily ruled out. Since Hg is highly volatile and its stable isotopes span a relatively large mass range (196–204), it may undergo mass-dependent isotopic fractionation during evaporation and condensation processes. Furthermore, the addition of Hg from distinct nucleosynthetic environments would also alter its isotopic composition. Since ^{198}Hg is only produced in s-process nucleosynthesis, the addition of s-process material would primarily result in an apparent overabundance of this isotope and an underabundance of the r-process-only isotope ^{204}Hg (Arlandini et al., 1999). Reactions

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during r-process nucleosynthesis would deplete the abundances of the light isotopes ^{196}Hg and ^{198}Hg , but ^{200}Hg , ^{201}Hg and ^{204}Hg would be enhanced relative to ^{202}Hg and normal isotopic composition. Heymann and Liffman (1986) were the first to investigate the effects of mini r-processing (neutron burst) on the isotopic composition of Hg. The latest calculations (Meyer et al., 2000, 2001) show that this process destroys ^{196}Hg and ^{198}Hg , while isotopes ^{199}Hg - ^{204}Hg are produced, with the relative enrichment increasing with mass when compared to terrestrial abundances. Hence, if there truly is a variation in the isotopic composition of meteoritic Hg as a result of either mass fractionation or nuclear processes, then it should be apparent in the abundances of the other five stable isotopes. Recent advances in the ability of ICP-MS to simultaneously measure low concentrations (pg/g levels) and isotopic abundances of Hg (Klaue et al., 2000; Klaue and Blum, 2000) led us to investigate the abundance and isotopic composition of Hg in meteorites. We began with two well-studied carbonaceous chondrites: Murchison (CM) and Allende (CV), both of which reportedly contain isotopically anomalous Hg.

2. PREVIOUS WORK

There are only a limited number of studies on the fractionation of Hg isotopes in natural systems. Haeffner (1953) investigated isotopic fractionation of Hg in an electric field and reported large (3%) variations. Obolenskii and Doilnitsyn (1976) reported fractionation in a series of cinnabar samples, suggesting that volcanic and hydrothermal deposits are enriched in heavy Hg isotopes relative to epithermal or telethermal deposits. However, their data lack the necessary precision to be confident of this conclusion (Koval et al., 1977; Kuznetsov and Obolenskii, 1980). Jackson (2001) recently reported non-mass-dependent fractionation of several Hg isotope ratios in lake sediments and associated organisms. These data were obtained by continuous-flow, cold-vapor-generation, quadrupole ICP-MS but are of low precision ($\sim 3\%$). The proposed mechanism leading to isotopic variability involves a series of complicated, kinetic fractionation steps to explain the distinct isotopic patterns, which seems unlikely given the high mass of Hg isotopes. Klaue et al. (2000) did, however, find mass-dependent fractionation in cinnabar and native Hg samples from a series of mines from throughout the world.

There have been three studies of Hg in Murchison (Jovanovic and Reed, 1976a, 1980; Kumar and Goel, 1992) and seven of Allende (Ozerova et al., 1973; Jovanovic and Reed, 1976a, 1980; Nier and Schlutter, 1986; Kumar and Goel, 1992; Ebihara et al., 1998; Kumar et al., 2001). The results of these studies are summarized in Tables 1 and 2. All but two of the studies rely on NAA for both abundance and isotopic measurements. Ozerova et al. (1973) used wet chemical techniques to measure the bulk abundance of Hg in Allende but did not carry out any isotopic measurements. Nier and Schlutter (1986) performed a gas-source mass spectrometry study of all seven Hg isotopes in bulk Allende and reported no detectable isotopic anomalies within their $\sim 20\%$ uncertainty. The bulk abundances of Hg that have been reported in the references above range from 48 to 494 ng/g in Murchison and from 5.8 to 10020 ng/g in Allende.

The reported isotopic anomalies were measured in thermal-

Table 1. Bulk Hg Abundances in Murchison and Allende

Bulk Hg Abundance (ppb)	Technique	Reference
Murchison		
>48	NAA	Jovanovic and Reed 1976a
65	NAA	Jovanovic and Reed 1980
494	NAA	Kumar and Goel 1992
294 ± 15	SC-ICP-MS	This work
Allende		
60	Wet Chemical	Ozerova et al 1973
35	NAA	Jovanovic and Reed 1980
37	NAA	Jovanovic and Reed 1980
32	NAA	Jovanovic and Reed 1980
9	NAA	Jovanovic and Reed 1980
976	NAA	Goel and Thakur 1989
189	NAA	Goel and Thakur 1989
2313	NAA	Goel and Thakur 1989
10020	NAA	Kumar and Goel 1992
4080	NAA	Kumar and Goel 1992
548	NAA	Kumar and Goel 1992
715	NAA	Kumar and Goel 1992
789	NAA	Kumar and Goel 1992
349	NAA	Kumar and Goel 1992
190	NAA	Kumar and Goel 1992
215	NAA	Kumar and Goel 1992
498	NAA	Kumar and Goel 1992
671	NAA	Kumar and Goel 1992
1212	NAA	Kumar and Goel 1992
16.4	NAA	Ebihara et al. 1998
18.3	NAA	Ebihara et al. 1998
30 ± 1.5	SC-ICP-MS	This work

release experiments in which the meteorites were heated to temperatures as high as 1200°C . The anomalous component comprised as much as 20% of the total Hg in Murchison and 85% of the total Hg in Allende. The temperature at which the anomalous Hg was released varied from 100 to 600°C for Murchison and from 100 to 1200°C for Allende. The anomalous fractions have $\delta^{196}\text{Hg}$ values that range from -260% to $+440\%$ in Murchison and from -620% to $+540\%$ in Allende (Fig. 1).

$$\delta^{196}\text{Hg} \equiv \left[\frac{(^{196}\text{Hg}/^{202}\text{Hg})_{\text{sample}}}{(^{196}\text{Hg}/^{202}\text{Hg})_{\text{standard}}} - 1 \right] * 1000 \quad (1)$$

We calculated the bulk $\delta^{196}\text{Hg}$ values ($\delta^{196}\text{Hg}_{\text{bulk}}$) for both meteorites using the fraction of anomalous Hg (X_{anom}) and the $\delta^{196}\text{Hg}$ values in the anomalous portion ($\delta^{196}\text{Hg}_{\text{anom},i}$) via the equation:

$$\delta^{196}\text{Hg}_{\text{bulk}} \equiv \sum_i \delta^{196}\text{Hg}_{\text{anom},i} \times X_{\text{anom},i} \quad (2)$$

where i represents the total number of heating steps in which anomalous components were released. The bulk isotopic composition ($\delta^{196}\text{Hg}$) calculated in this manner varies between -16 and $+99\%$ for Murchison and -153 to -1% for Allende (Table 2).

The wide range in meteoritic Hg measurements results from one of several possibilities: a) heterogeneous distribution of Hg of varied isotopic composition throughout all classes of mete-

Table 2. Hg Isotopic Anomalies Reported in Murchison and Allende

Total Hg (ppb)	T _{release} (°C)	Anomalous Hg (ppb)	X _{anom} ^a	δ ¹⁹⁶ Hg _{anom} (‰)	δ ¹⁹⁶ Hg _{bulk} (‰)	Reference
Murchison						
>48	600	4.96	0.103	-153	-16	Jovanovic and Reed 1976a
494	100	98	0.198	-260	99	Kumar and Goel 1992
	200	191	0.387	440		
	400	68	0.138	-140		
Allende						
32 ^b	600	0.14	0.044	-342	-16	Jovanovic and Reed 1976a
	1200	1.4	0.004	-145		
57 ^b	600	0.79	0.014	-329	-8	Jovanovic and Reed 1976a
	1200	0.3	0.005	-571		
5.8 ^b	1200	1.45	0.250	-310	-78	Jovanovic and Reed 1976a
241 ^b	600	2.2	0.009	-145	-1	Jovanovic and Reed 1976a
34 ^c	250	20	0.588	-213	-153	Jovanovic and Reed 1976a
	250	5.6	0.165	-167		
976 ^d	150	976	nr	-620		Goel and Thakur 1989
189 ^d	250	189	nr	540		Goel and Thakur 1989
10020 ^d	100	1190	0.119	-120	-34	Kumar and Goel 1992
	300	1140	0.114	-170		
4080 ^d	100	520	0.127	-166	-103	Kumar and Goel 1992
	300	400	0.098	-200		
	400	690	0.169	-232		
	500	490	0.120	-190		
548 ^d	200	438	0.799	-110	-108	Kumar and Goel 1992
	300	95	0.173	-190		
	400	15	0.027	460		
715 ^d	300	117	0.164	-260	-43	Kumar and Goel 1992
789 ^d	300	188	0.238	-250	-60	Kumar and Goel 1992
349 ^d	300	80	0.229	-440	-85	Kumar and Goel 1992
	400	23	0.066	240		
190 ^d	300	31	0.163	-150	-24	Kumar and Goel 1992
215 ^d	300	50	0.233	-120	-28	Kumar and Goel 1992
498 ^d	300	60	0.120	-170	-20	Kumar and Goel 1992
671 ^d	300	90	0.134	-280	-33	Kumar and Goel 1992
	400	26	0.039	120		
1212 ^d	400	820	0.677	-200	-135	Kumar and Goel 1992

^a mass fraction of total Hg that had anomalous isotopic composition

^b matrix

^c chondrules

^d successive distillates of bulk meteorite

orites, b) contamination of meteorite samples by terrestrial sources (Somayajulu, 1975), or c) systematic errors in the analytical techniques. Kumar et al. (2001) suggest that previous measurements may have failed to consider the interference between ²⁰³Hg and ⁷⁵Se for NAA, resulting in the wide variability in reported meteoritic Hg abundances and isotopic ratios. Accurate, precise, and contamination-free measurements of Hg isotopes in meteorites are needed to resolve these issues.

The only Hg-bearing phase identified in meteorites is HgS, reported from both the Allende (CV) and Mighei (CM) chondrites. In Allende, the HgS occurs as small (~5 μm) grains at the base of troilite-pentlandite rims in sulfide-rich nodules embedded in an unusual xenolith (Kurat et al., 1989). The HgS in Allende is interpreted by Kurat et al. (1989) as the product of low-temperature condensation in the solar nebula under a high sulfur fugacity. This formation mechanism is consistent with the Hg condensation model presented in Laurretta et al. (1999) and observations of sulfide mineral assemblages in Allende matrix and chondrules (e.g., Blum et al., 1989).

The HgS grains in Mighei are associated with heavily altered chondrules that also contain phyllosilicates, calcite, and P-rich sulfides (Laurretta et al., 1999). In contrast to the HgS in Allende, the grains in Mighei are probably the products of aqueous alteration on the CM chondrite parent body. Thus, low-temperature processes in the solar nebula and on meteorite parent asteroids are reflected in meteoritic Hg chemistry. By investigating Hg abundances, isotopic compositions, and thermal-release profiles, we hope to better constrain the conditions under which these processes occurred.

3. EXPERIMENTAL METHODS

3.1. Samples and Preparation Techniques

All meteorites were obtained from the Center for Meteorite Studies at Arizona State University. Three different samples of Murchison and Allende were used to check for variability across individual specimens. Clean samples were obtained by removing the fusion crust with a high-speed diamond sanding wheel and immediately isolating the interior pieces. These pieces were further disaggregated under clean-

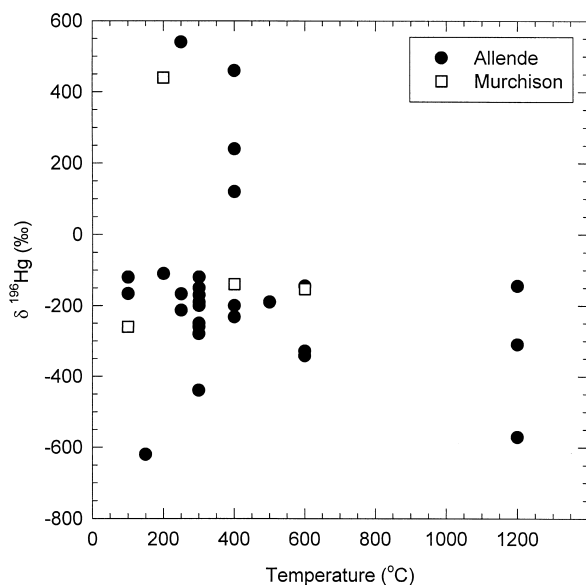


Fig. 1. Graph showing the magnitude of anomalies in the $^{196}\text{Hg}/^{202}\text{Hg}$ ratio reported in the literature for Murchison and Allende as a function of release temperature. The $\delta^{196}\text{Hg}$ values range from -620 to $+540$ ‰ for Allende and from -260 to $+440$ ‰ for Murchison. Most of the anomalous Hg is released from the meteorites between 150 and 600°C.

room conditions to obtain uncontaminated material from the center of each sample. For bulk analyses we digested a total of ~ 15 g of Allende and ~ 2.5 g of Murchison.

The samples were digested using two techniques. The recovery rates for both methods were tested using standard reference materials (see below). The first method is based on low-pressure, low-temperature acid dissolution with a mixture of HF/HNO₃/HCl in tightly capped 15 mL PFA vessels (Saville) at temperatures of 80 to 90°C. The Hg recovery rates were around 98 to 99%. The residual mass, which consisted mostly of graphite, kerogen, and complex fluorides, was further dissolved using HClO₄ but resulted in negligible Hg yields. Samples were also prepared using high-pressure, microwave-accelerated acid dissolution (e.g., Kingston, 1997). The closed-vessel system (MARS5, XP1500 vessels, CEM Inc., NC) prevents the loss of volatile species such as Hg and allows the complete dissolution of even black carbon fractions under aggressive conditions (220°C, 70 bar) in a mixture of HF/HCl/HNO₃. Recovery rates of the standard reference materials again proved to be excellent (93 to 100%). The microwave-accelerated digestion left white residues of complex fluorides, which were further treated with boric acid at 200°C and 40 bars. Again, no Hg was detected in the residue.

We preconcentrated both Allende and Murchison digests by employing sequential cold-vapor (Hg^0) generation. The preconcentration was essential for the multi-collector (MC) ICP-MS isotopic analyses because of sensitivity limitations. The acidic samples were mixed in a batch reactor with alkaline 1% sodium borohydride solution (Klaue and Blum, 1999), and the released Hg vapors were condensed in a cold trap. Although the cold-vapor generation with sodium borohydride has the disadvantage of slightly higher background levels compared to the alternative HCl/Sn(II) cold-vapor generation reagent, it offers significantly better recovery rates for the batch release of Hg out of the strong acid-digestion mixtures. The Hg in the cold trap was then dissolved in concentrated HNO₃ and diluted with water for the final continuous-flow, cold-vapor generation MC-ICP-MS analyses (see below). Hg laboratory standards with previously well-characterized isotopic compositions were processed in the identical way from digestion to final isotopic analyses. No significant isotopic fractionation over the course of the digestion and preconcentration procedure was observed.

The final preparation step reduced all Hg^{2+} to Hg^0 using Sn(II) and provided a highly efficient sample-introduction method for the isotopic analyses. The isobaric interferences of ^{196}Pt , ^{198}Pt , and ^{204}Pb , as well as molecular interferences from tungsten oxides (WO), metal hydrides (TiH), and tin argides (ArArSn) were eliminated by the chemistry of the reduction reaction and the design of the gas-liquid separator. Hence, there was no need for chromatographic separations and possible problems with Hg memory effects or blanks in the separation procedure were avoided. The transport efficiency of Hg through the gas-liquid separator at the applied concentration levels was greater than 99% and, thus, did not cause any measurable isotopic fractionation.

3.2. Standards

We employed standard reference materials (SRM's) for external quality control of the bulk chemical abundance of Hg in the meteorite samples. There are only a few geological reference materials available with suggested Hg concentrations. We used two certified SRM's from the Geological Survey of Japan: JG-1 Granodiorite (16.5 ppb Hg) and JB-1 Basalt (4.78 ppb Hg). Both Hg concentration values are labeled "preferable" as opposed to the truly certified or at least "recommended" values. Nevertheless, the excellent agreement of those values with our measurements suggested that they are correct within our analytical precision (3–5%). Calibration was performed by the standard addition method with NIST traceable standard materials from High Purity Standards (P.O. Box 80609, Charleston, SC 29416).

As listed in Table 3, reported terrestrial Hg isotopic abundances show significant discrepancies (Zadnik et al., 1989). The most recent determination of the isotopic composition of terrestrial Hg is by Zadnik et al. (1989), and their values are currently accepted by IUPAC (Rosman and Taylor, 1998). However, there are significant differences between the data of Zadnik et al. (1989) and those of Nier (1950), which were the previously accepted IUPAC values (Holden et al. 1984). Our ICP-MS results are a much closer match to the data of Nier (1950) for most isotope ratios except for $^{196}\text{Hg}/^{202}\text{Hg}$, which he did not measure. Klaue and Blum (2000) discuss the problem of the true Hg isotopic abundance and the range of values obtained by ICP-MS and gas-source mass spectrometry.

Table 3. Isotopic Composition of Terrestrial Hg

Ratio	IUPAC 1983 ^{a,c}	IUPAC 1997 ^{b,e}	SC-ICP-MS ^{c,e}	SC-ICP-MS ^{d,e}	MC-ICP-MS ^{d,e}
196/202	0.00491 (15)	0.00514 (1)	0.00517 (4)	0.00519 (1)	0.005183 (1)
198/202	0.3362 (3)	0.3338 (4)	0.3377 (3)	0.3386 (3)	0.33726 (2)
199/202	0.5650 (5)	0.5650 (5)	0.5709 (3)	0.5710 (3)	0.56931 (3)
200/202	0.7761 (5)	0.7734 (8)	0.7796 (4)	0.7790 (4)	0.77767 (6)
201/202	0.4437 (6)	0.4414 (4)	0.4438 (2)	0.4450 (2)	0.44271 (2)
204/202	0.2299 (2)	0.2299 (2)	0.2298 (1)	0.2304 (1)	0.22917 (2)

^a based on results of Nier (1950)

^b from Zadnik et al. (1989)

^c average procedural blank (5–10 pg/g)

^d from analyses of the Almaden ore and product diluted to 0.9 ng/g

^e 2 SE on last digit(s) given in parentheses

For the analysis of the isotopic compositions we used purified elemental Hg from the Almaden mine in Spain as a relative “zero” fractionation standard. This mine is the largest known deposit of cinnabar on Earth (Hernandez et al., 1999). We measured the Hg isotopic compositions of several cinnabar samples from different deposits within the mine and the purified elemental Hg product. Within our analytical precision of 0.05 ‰ we found no measurable difference in their isotopic compositions. Therefore, we adopted the Almaden Hg as a reasonable isotopic composition for terrestrial Hg.

3.3. Measurement Techniques and Data Reduction

Bulk chemical abundance and isotopic composition analyses were performed by cold-vapor-generation ICP-MS with two different instruments. For the concentration analyses, an ELEMENT (Finnigan MAT, Bremen, Germany) single collector (SC), magnetic-sector ICP-MS was employed. This instrument provides superior sensitivity for the direct analyses of Hg in acid dissolution samples (Chen et al., 2000). Relative detection limits are on the order of 0.1 ng/kg, and absolute detectable amounts are ~100 fg. Based on the blanks for the digestion procedures, the sensitivity exceeded the expected concentration levels in the meteorites by at least a factor of 500. The machine background (Ar, air entrainment, internal sources, etc.) was negligible compared to the procedural background (<1%). The continuous-flow, cold-vapor-generation apparatus consisted of a modification of the gas-liquid separator described by Klaue and Blum (1999). A solution of 3% Sn(II)Cl₂ in 1 mol/L HCl was used to reduce Hg²⁺ to Hg⁰. The sample solution and the reducing Sn solution were mixed online and pumped into the gas-liquid separator. The Hg “cold” vapor separated from the liquid and was swept into the plasma of the ICP-MS by a continuous flow of argon. Because of the high sensitivity of the SC-ICP-MS, no preconcentration was necessary.

For the bulk isotopic analyses, we coupled the gas-liquid separator to a P54 (VG Elemental Inc., UK) MC-ICP-MS. This instrument is equipped with nine Faraday cups so that all seven Hg isotopes and two isotopes of an internal standard (²⁰³Tl and ²⁰⁵Tl) can be analyzed simultaneously. The use of Tl from the NIST SRM 997 “Isotopic Standard for Thallium” as an internal standard has been widely employed for the analyses of Pb isotopes (Hirata, 1996; Rehkaemper and Halliday, 1998; White et al., 2000). Tl cannot be introduced by cold-vapor generation. Instead, a micro-concentric nebulizer (MCN-T1, CETAC Technologies, Omaha, NE) was used with a membrane desolvation unit (ARIDUS, CETAC). This technique provides a dry thallium nitrate aerosol, which is mixed with the Hg cold vapor after the gas-liquid separator. The introduction of a dry aerosol maintains the high sensitivity of the P54 by reducing the kinetic-energy spread of the M⁺ ions in the plasma and minimizes oxide or hydride molecular interferences in the plasma (Appelblad et al., 2000).

The measured Hg isotope ratios were corrected for instrumental mass fractionation using the internal Tl standard and employing a power law function (Hirata, 1996; Habfast, 1998; Rehkaemper and Halliday, 1998; Marechal et al., 1999; White et al., 2000). Despite the problems associated with using a simple power law mass-fractionation correction for MC-ICP-MS (Rehkaemper and Halliday, 1998; Marechal et al., 1999; White et al., 2000), our values agree well with previously reported data. The results obtained by the SC-ICP-MS (ELEMENT) and the MC-ICP-MS (P54) also match within the precision range of both methods (see Table 3). This agreement shows that our MC-ICP-MS method is capable of determining Hg isotopic compositions accurately and precisely. The relative differences in the isotopic compositions of different samples can be resolved down to the ±0.05 ‰ level, given sufficient amounts (>1 μg) of Hg for the analysis. Because of the multiple Faraday-cup detector array, sensitivity is the major limitation for the analysis of samples with low Hg levels, especially for ¹⁹⁶Hg. Typically, about 300 ng of Hg are necessary to obtain optimal internal precision levels for all isotopes. Isotopic analyses in the range of 0.1 to 0.2 ‰ internal precision levels are possible with ~100 ng total Hg.

Thermal release profiles were determined online by coupling a programmable tube furnace to the SC-ICP-MS. The furnace consisted of a 22-mm (ID), thin-wall, quartz-glass tube, which was wrapped with high-temperature, ceramic-beaded, Ni-Cr-V resistance wire over a length of ~10 cm. In the middle of the heated area a 6-mm ID quartz

Table 4. Isotopic Composition of Meteoritic Hg

Isotope Ratio	Allende*	Murchison*
SC-ICP-MS Analyses		
196/202	0.00520 (2)	0.00518 (1)
198/202	0.3390 (3)	0.3386 (3)
199/202	0.5713 (5)	0.5706 (5)
200/202	0.7796 (6)	0.7787 (6)
201/202	0.4452 (4)	0.4448 (3)
204/202	0.2304 (2)	0.2305 (2)
MC-ICP-MS Analyses		
196/202	0.005181 (4)	0.005184 (3)
198/202	0.33724 (7)	0.33727 (8)
199/202	0.56928 (14)	0.56934 (12)
200/202	0.77760 (15)	0.77731 (15)
201/202	0.44275 (9)	0.44269 (8)
204/202	0.22914 (5)	0.22920 (6)

* 2 SE on last digit(s) given in parentheses

glass thermowell was inserted holding a type-K thermocouple. Temperature control and data logging were performed with a Digi-Sense Advanced Temperature Controller (Cole Parmer, Vernon Hills, IL, USA). The temperature probe was calibrated at 0 and 100°C. Small (1–100 mg) aliquots of powdered meteorites and standards were placed in glazed porcelain boats (5 × 7 mm) in the isothermal region of the furnace. The samples were heated from room temperature to 900°C along a linear temperature ramp over 25 min. This method allowed us to determine when distinct phases were releasing Hg. Because of the high sensitivity of the ELEMENT and the low background levels, we were able to achieve detection limits of less than 1 fg Hg. None of the published thermal decomposition procedures for meteorites provided this information. This experiment was performed three times for each meteorite. In the first and second runs we used low mass-resolution mode ($M/\Delta M_{(5\%)}=400$) to measure the abundance of each Hg isotope and the intensity of Hg and a suite of other trace elements (including ⁸²Se), respectively. During the third run we used the medium mass-resolution mode ($M/\Delta M_{(5\%)}=4000$) to simultaneously monitor the release of ¹⁹⁶Hg, ¹⁹⁸Hg, ³²S, ¹⁶O, and ³⁵Cl.

We performed thermal release experiments for a variety of materials including: 1) synthetic Hg compounds [HgCl₂, black HgS (metacinnabar), HgSe, HgO, and HgSO₄]; 2) natural Hg-bearing material [red HgS (cinnabar) from Almaden, ZnS (sphalerite) from the Tri-State Mississippi Webb City mine, and the JG-1 granodiorite and JB-2 basalt SRMs from the Geological Survey of Japan]; and 3) Hg⁰ adsorbed on different materials [SiO₂, Au, heat-treated Allende (baked at 900°C under Ar), steel, and SiC]. Small amounts of the pure Hg compounds were mixed with SiC such that the Hg abundances were ~100 ppb, similar to the concentrations in the meteorite samples. For the experiments with adsorbed Hg⁰, the material was baked at 900°C then placed inside a sealed beaker with a saturated Hg⁰ atmosphere for one hour. The material was then subjected to the same thermal profile as the meteorite samples.

4. RESULTS

4.1. Bulk Abundances and Isotopic Compositions

The bulk Hg concentrations for Allende and Murchison are 30 ± 1.5 ng/g and 294 ± 15 ng/g, respectively. These values were determined after both low-pressure and microwave-accelerated, high-pressure dissolution with no significant difference between the two methods and independent sample aliquots. Analyses of Allende and Murchison samples by SC- and MC-ICP-MS revealed only small differences in the isotopic compositions compared to the Almaden reference sample. The

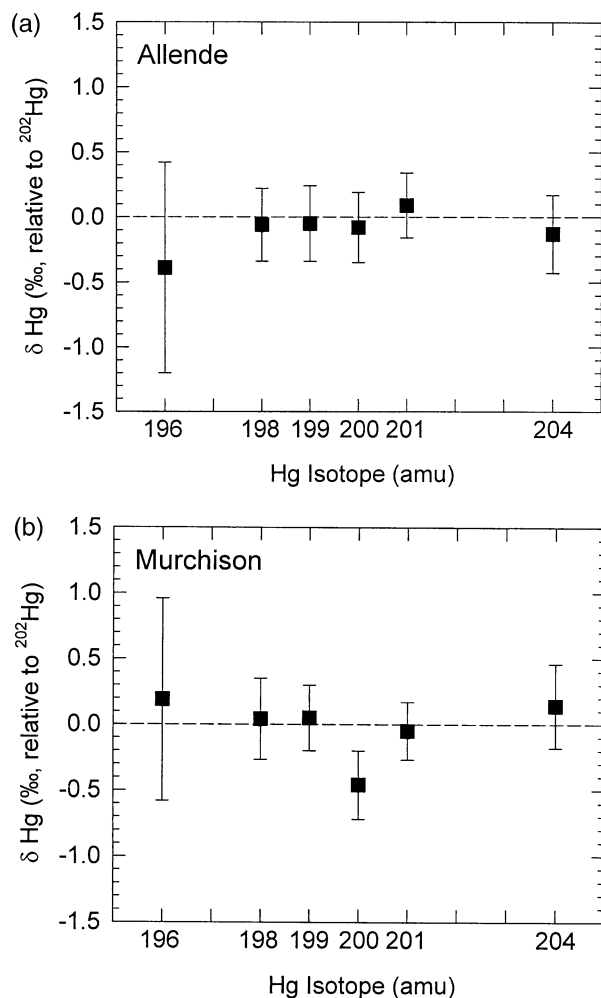


Fig. 2. Graphs showing the bulk isotopic composition of Hg in Allende (2a) and Murchison (2b). The error bars reflect the most conservative error based on the SE of the standard and sample analyses (see text). There are only small differences between the isotopic compositions of the meteorites (circles) and our standard (dashed lines).

results for the isotopic analyses by SC- and MC-ICP-MS for the meteorite samples are summarized in Table 4. The reported standard error (SE) of the meteoritic isotope ratios reflects only the internal precision of the 75 replicate cycles (5 blocks, 15 cycles, 5s integration time) that were possible with the limited amounts of Hg available (150–250 ng) from the Allende and Murchison samples. The SE for the reference standard represents the external precision of 15 runs (5 blocks, 20 cycles, 5s integration time) of the Almaden sample before and after the meteorite sample runs. The abundances of the Hg isotopes (relative to the terrestrial standard) as determined by MC-ICP-MS are shown in Figures 2a and b. The absolute uncertainties shown in Figure 2a and 2b reflect the maximum and minimum δ values calculated by adding or subtracting the 2SE of the absolute ratios of the reference (Table 3) and the samples:

$$\delta_{\max} = (((R_{\text{sample}} + 2\text{SE}) / (R_{\text{standard}} - 2\text{SE})) - 1) * 1000 \quad (3)$$

$$\delta_{\min} = (((R_{\text{sample}} - 2\text{SE}) / (R_{\text{standard}} + 2\text{SE})) - 1) * 1000 \quad (4)$$

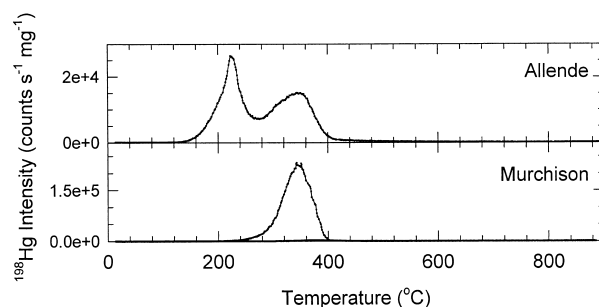


Fig. 3. Graphs showing the thermal release profile of ^{198}Hg in both Allende and Murchison. Allende releases Hg at two distinct temperatures, 225 and 344 $^{\circ}\text{C}$. At each temperature $\sim 50\%$ of the total Hg is released. Murchison releases Hg only at 343 $^{\circ}\text{C}$. For both meteorites there is no indication of significant Hg release above $\sim 450^{\circ}\text{C}$.

The application of this simple method instead of a formal error propagation calculation is the most conservative way to calculate the uncertainties associated with our measurements (Klötzi 1992). The bulk isotopic compositions of both meteorite samples are identical to our standard for most isotope ratios, within the analytical uncertainty. The only exception is the $^{200}\text{Hg}/^{202}\text{Hg}$ ratio for Murchison, with a δ value of -0.45‰ . Neither data set displays any significant mass-dependent fractionation trend.

4.2. Thermal Release Profiles

We monitored the amount of Hg released from Allende and Murchison as a function of temperature (Fig. 3). The patterns for the two meteorites are distinctly different. This difference is believed to be real, since the release patterns for both Murchison and Allende were identical in five replicate experiments. The Hg release pattern for Allende has two maxima, at 225 $^{\circ}\text{C}$ and 344 $^{\circ}\text{C}$. All the Hg in Murchison is released in a single peak at 343 $^{\circ}\text{C}$. There is no indication of Hg release above $\sim 500^{\circ}\text{C}$ for either meteorite.

We also determined the isotopic ratios (relative to ^{202}Hg) of the Hg released from Allende (Fig. 4) and Murchison (Fig. 5) as a function of temperature. The isotopic compositions are constant within the precision of the SC-ICP-MS measurement technique ($\sim 5\text{--}30 \text{‰}$ depending on the isotope ratio) at all temperatures where significant Hg was released. The isotope ratios measured during thermal release are of lower precision than the bulk meteorite values obtained by SC- and MC-ICP-MS. The slight deviations from terrestrial ratios are not significant, and there is no evidence of the large variations reported in the NAA studies for distinct fractions released at various temperatures (see Table 2).

The contrast between the thermal release patterns of Hg from Allende and Murchison suggests that there is a significant difference in the Hg chemistry of the two meteorites. The thermal release experiments provide insight into the reasons for this variation [Figure 6 (Hg compounds), Figure 7 (natural materials), and Figure 8 (adsorbed Hg)]. The release patterns for the different Hg species are all unique. The peak release temperatures range from 131 $^{\circ}\text{C}$ (HgCl_2) to over 800 $^{\circ}\text{C}$ (Hg in ZnS, JB-2). The patterns for adsorbed Hg are equally complex. Even chemically uniform compounds have multiple peaks in

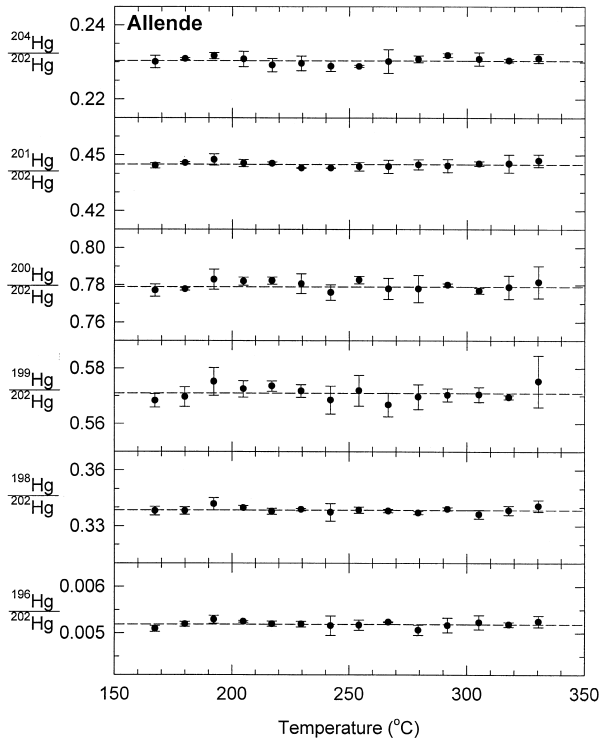


Fig. 4. Graphs showing the variation in Hg isotopic ratios released from Allende at increasing temperature. The averages of all six ratios are identical to terrestrial values. The largest variations are observed in $^{196}\text{Hg}/^{202}\text{Hg}$, which scatter by as much as $\pm 20\%$ around the average. However, all values are identical to those in the standard, within the precision of the method.

their Hg thermal release profiles (e.g., SiO_2 , Au, and SiC). Biester and Nehrke (1997) published thermal release patterns of metacinnabar and cinnabar, which are generally consistent with our profiles except for small differences in temperature ranges. The discrepancies in Hg release temperatures can probably be attributed to differences in the applied temperature gradient or the absolute calibration.

In separate experiments, the release patterns of Hg and S were determined simultaneously, allowing us to further constrain the chemistry of Hg in these two meteorites (Fig. 9). The low-temperature Hg-release peak for Allende does not correlate with that of S. However, the second peak correlates with a S peak. The peak in the Murchison Hg-release pattern also correlates with S. There is another, higher temperature peak (425°C) in the Allende S-release pattern that is not associated with Hg release. For both meteorites, most of the S is released at higher temperatures.

We determined the thermal release patterns of Se from both Allende and Murchison to evaluate the possibility of an interference of ^{75}Se with ^{203}Hg in the NAA studies. The Se release patterns for both meteorites are distinctly different (Fig. 10). Little Se is released from Allende at low temperatures. Most Se is released above 700°C , and the peak appears to be above 900°C , which represented the maximum temperature of our experiments. However, there are several small peaks in the Se release pattern for Allende at temperatures as low as 150°C .

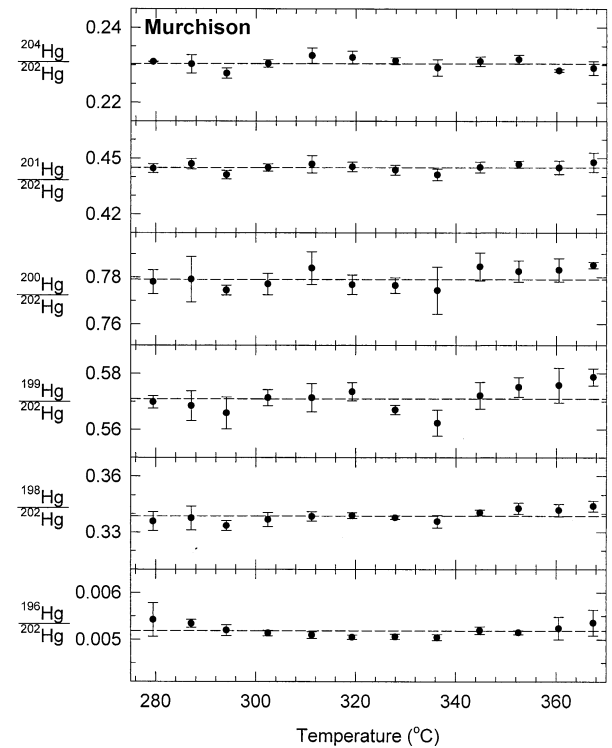


Fig. 5. Graphs showing the variation in Hg isotopic ratios released from Murchison at increasing temperature. The averages of all six Hg isotopic ratios are identical to terrestrial values. The largest variations are observed in the $^{196}\text{Hg}/^{202}\text{Hg}$, which scatter by as much as $\pm 30\%$ around the average. However, all values are identical to those in the standard, within the precision of the method.

Murchison, on the other hand, releases most Se at 410°C and has a broad second peak at 760°C .

5. DISCUSSION

5.1. Contamination

Contamination of the samples by terrestrial Hg is a concern that requires careful consideration (Somayajulu, 1975). We performed several experiments to determine the possible contribution of terrestrial Hg to our results. First, we compared the bulk Hg abundance from regions near the fusion crust with the interiors of the meteorites. The results from both regions are identical, suggesting either the samples are saturated with terrestrial Hg or that little Hg has diffused into the sample. To distinguish between these two possibilities, we also exposed a $\sim 1\text{cm}^3$ piece of Allende to a saturated Hg^0 atmosphere for 24 h and then performed the identical cleanup procedure as for the bulk abundance analyses. No difference in the extent or shape of the release pattern was observed.

A third experiment was performed to further investigate the possibilities of sample contamination. A powdered sample of Allende was baked at 900°C under Ar until it released all of its Hg and then exposed to Hg vapor for 24 h. The bulk of the meteorite is composed of relatively refractory material that should not be greatly affected by heating to this temperature. The thermal release profile of Hg from this sample was deter-

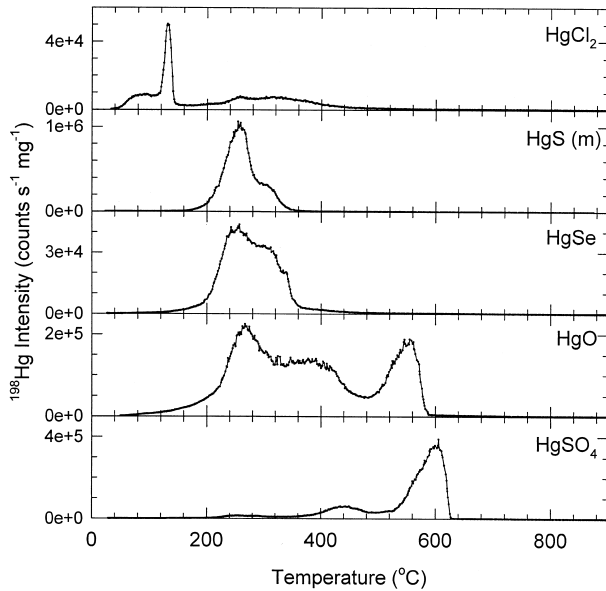


Fig. 6. A series of graphs showing the thermal release profiles of ^{198}Hg from a variety of Hg compounds. Each release profile is unique. The temperatures at which peak Hg release occurs vary from 131°C for HgCl_2 to 600°C for HgSO_4 . Comparison of these profiles to the meteoritic data constrains the mineralogy of Hg in Allende and Murchison.

mined (Fig. 8). The pattern is distinct from that of fresh Allende. The amount of Hg in the contaminated material is $\sim 10 \times$ that of the uncontaminated material. The onset of Hg release in the contaminated sample occurs $\sim 50^\circ\text{C}$ lower than from the fresh material, and the peak is lower and much broader than the first peak from the fresh samples (206°C vs. 225°C). There is no indication of a peak at 340°C from the contaminated sample. These differences show that our thermal analysis ICP-MS method can distinguish between meteoritic Hg and

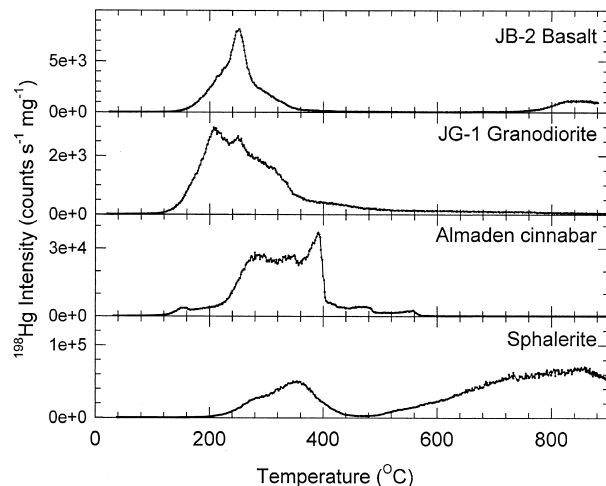


Fig. 7. A series of graphs showing the thermal release profiles of ^{198}Hg in natural samples. The profile of Hg released at low temperatures from Allende most resembles that from the basalt, suggesting that Hg in Allende may be adsorbed on silicate grains.

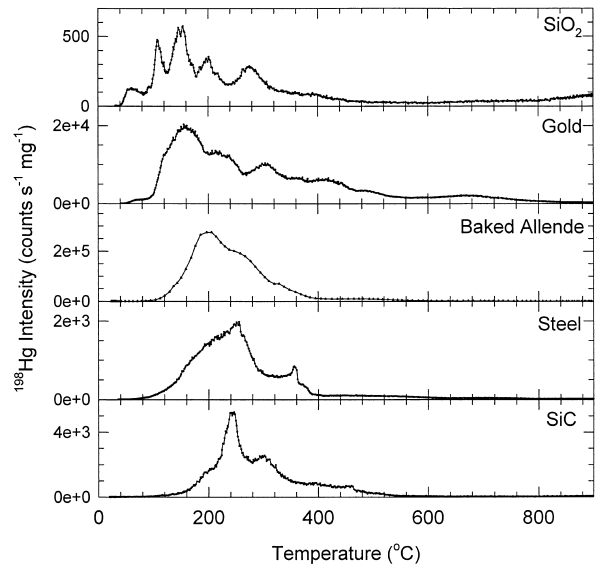


Fig. 8. A series of graphs showing the thermal release profiles of ^{198}Hg adsorbed on different materials. The temperature at which peak Hg release occurs is similar for all materials. However, the details of the profiles are all different.

terrestrial contamination. In addition, the distinctions between the two profiles suggest that contamination is not a major issue in the meteorite experiments.

5.2. Bulk Abundance of Hg

There have been numerous measurements of the bulk Hg abundance in Allende (Table 1). Measurements by Jovanovic and Reed (1976a, 1976b, 1980) and Reed (2000) gave Hg concentrations of 6, 32, 57, and 241 ppb for bulk or matrix samples; 9, 32, 35, and 37 ppb for bulk samples; and 37.5 and 34 ppb for Allende chondrules. Thus, six of their analyses gave Hg concentrations between 32 and 37 ppb. Ozerova et al. (1973) reported a bulk Hg concentration of 60 ppb. More recent studies (Goel and Thakur, 1989; Kumar and Goel, 1992) report

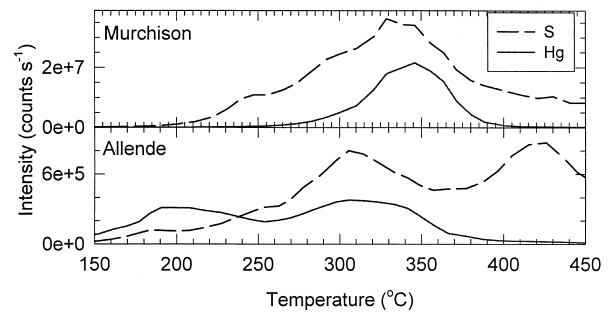


Fig. 9. Graphs comparing the release profiles of S and Hg from Allende and Murchison. The two elements have very similar release patterns for Murchison, suggesting that they are released from the same phase. The Hg released at low temperatures from Allende does not correlate with S release, but the higher temperature portion does. These data suggest that Hg is distributed in two different sites in Allende, one of which is most likely a sulfide.

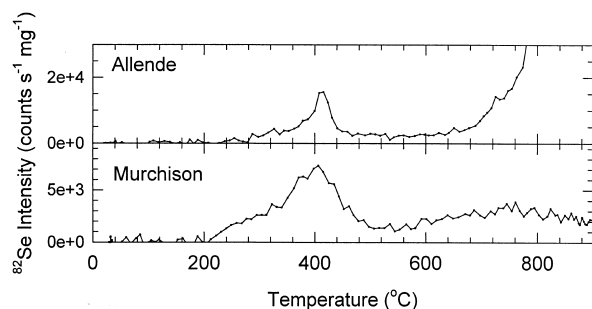


Fig. 10. Graphs showing the release patterns of ^{82}Se from Allende and Murchison. Allende releases most of its Se above $\sim 700^\circ\text{C}$. There is a small amount released around 400°C . Murchison, on the other hand, only has a peak in its Se release profile at $\sim 400^\circ\text{C}$. However, the amount of Se released at this temperature is insignificant compared to the amount of Hg, suggesting that a Se interference did not cause the anomalous Hg isotopic ratios reported previously.

much higher bulk abundances, with values ranging from 189 to 10020 ppb Hg. Finally, Kumar et al. (2001) report bulk Hg abundances of 16.4 and 18.3 ppb Hg in two different experiments. Repeated measurements in our laboratory consistently gave a bulk abundance of 30 ± 1.5 ng/g, in agreement with most of the earlier NAA studies.

The earliest bulk measurements of Hg in Murchison report abundances of >48 and 65 ppb Hg (Jovanovic and Reed, 1976a, 1980), respectively, whereas Kumar and Goel (1992) reported 494 ppb Hg. Our measurements fall between these two studies, with bulk Hg abundances in Murchison equal to 294 ± 15 ng/g. We observed a much larger variation in Murchison bulk Hg abundances compared to Allende, suggesting that Hg is heterogeneously distributed in Murchison and more evenly dispersed in Allende. However, the observed variations could also result from the smaller sample aliquots of Murchison (2g) used in the experiments.

5.3. Isotopic Composition of Meteoritic Hg

Previous studies of the isotopic composition of meteoritic Hg reported variations in the $^{196}\text{Hg}/^{202}\text{Hg}$ ratio. The reported anomalies are large and, in some cases, the anomalous component comprises a major fraction of the total Hg. Thus, the anomalous Hg isotopic compositions should be measurable in bulk samples (see Table 2). We determined the isotopic compositions of Hg in Allende (Fig. 2a) and Murchison (Fig. 2b). In almost every case there is no variation between the relative abundances of Hg isotopes in our standard and those in the meteorites. The only exception is the $^{200}\text{Hg}/^{202}\text{Hg}$ ratio in Murchison, which is lower than our reference value by more than the conservative maximum/minimum error. None of the nucleosynthetic mechanisms discussed above would be expected to selectively deplete this isotope (Inghram et al., 1947; Heymann and Liffman, 1986; Bao et al., 2000). Further measurements are needed to confirm the significance of this small isotopic difference, which falls only slightly outside of the analytical uncertainty.

The bulk isotopic composition measurements constrain the abundance and magnitude of an isotopically anomalous Hg component (Fig. 11). The anomalies reported in the $^{196}\text{Hg}/$

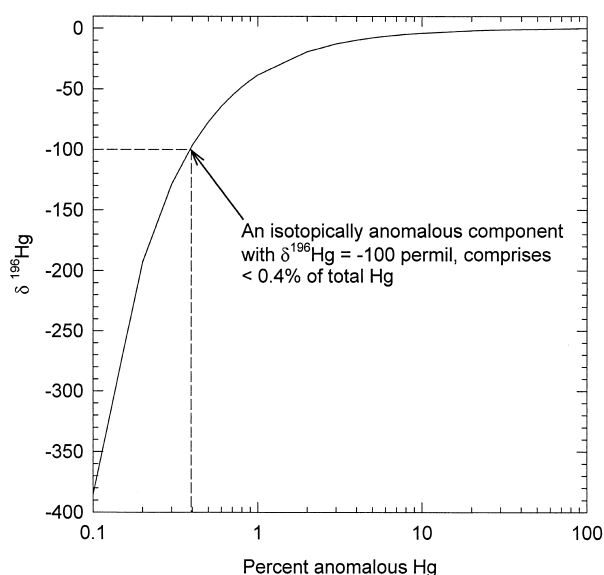


Fig. 11. Graph showing the maximum percent of anomalous ^{196}Hg in Allende based on our bulk isotopic composition measurement. The bulk sample has a $\delta^{196}\text{Hg}$ value of -0.4 ‰. Thus, if the anomalous component has $\delta^{196}\text{Hg} = -100$ ‰, it can only comprise 0.4% of the total Hg.

^{202}Hg ratio for bulk Allende in the literature range from -620 ‰ to $+540$ ‰. The bulk isotopic composition ($\delta^{196}\text{Hg}$) of Allende is -0.4 ± 0.8 ‰. Thus, an anomalous component with a $\delta^{196}\text{Hg}$ value of -100 ‰ can at most comprise 0.4% of the total Hg in Allende. If the anomalous component has a $\delta^{196}\text{Hg}$ of -620 ‰, the largest anomaly reported, it comprises less than 0.1% of the total Hg.

We attempted to isolate anomalous components by monitoring the isotopic composition of Hg during the thermal release experiments. In the previous studies, most of the anomalous Hg was released between 200° and 400°C (Fig. 1). This is the same temperature range in which we observed the majority of Hg release from these meteorites (Fig. 3). Thus, any anomalous component that was released from the meteorites at these temperatures would be easily discernable in our experiments. The isotopic variations of meteoritic Hg released from each meteorite as a function of temperature are shown in Figures 4 (Allende) and 5 (Murchison). The ratio of each Hg isotope relative to ^{202}Hg is constant and equal to terrestrial values within the precision of our technique. The $\delta^{196}\text{Hg}$ values vary by ± 20 ‰ in Allende and by ± 30 ‰ in Murchison. The other ratios vary by less than ± 10 ‰ from our standard. There is no obvious anomalous component, especially of the magnitude reported in previous studies.

5.4. Thermal Release Profiles and the Distribution of Hg

Perhaps the most striking result of this study is the difference in the Hg thermal release profiles from Allende and Murchison. This contrast suggests that there is a significant difference in the Hg chemistry of the two meteorites. By comparing the peak Hg release temperatures from a variety of pure Hg compounds, natural samples, and adsorbed elemental Hg to the meteorite

patterns, we can place constraints on the Hg-bearing phases. Since HgCl_2 releases most of its Hg at 131°C , we can eliminate it as a host phase. Likewise, HgO , HgSO_4 , and ZnS have significant Hg release above 500°C , suggesting that they do not contribute to the meteorite release patterns. We consider it unlikely that SiO_2 , Au, or SiC are present in the meteorites in sufficient abundance to account for the observed release patterns. The patterns of Hg release from the meteorites are broadly consistent with our experiments using metacinnabar, HgSe , cinnabar, basalt, and steel. It is, of course, also possible that Hg is contained in a phase that we did not investigate.

Monitoring the release of other elements (O, S, Cl) along with Hg helped us to further constrained the meteoritic Hg-bearing phases. It is apparent from the Allende release profiles that the first peak is not associated with a sulfide. Instead, the shape and structure of the peak closely resembles that of Hg released from basalt (Fig. 7). This similarity suggests that the Hg released at 225°C from Allende is physically adsorbed on some component in the meteorite matrix. The peaks at $\sim 340^\circ\text{C}$ in the Hg release patterns from both meteorites correlate with S release. This correlation suggests the material that releases Hg at 340°C contains abundant S, consistent with observations of HgS grains in CV (Kurat et al., 1989) and CM (Lauretta et al., 1999) chondrites.

On the basis of our experiments, we conclude that Hg is distributed between two different chemical sites in Allende. Some of the Hg is likely adsorbed on mineral surfaces and is released at low temperatures, whereas the rest occurs as HgS . Murchison does not contain any detectable adsorbed Hg. Instead, essentially all Hg appears to occur as HgS . The difference in the Hg chemistry between Allende and Murchison may reflect the low-temperature alteration history of these two meteorites. Murchison has experienced much more intense aqueous alteration. This process probably mobilized adsorbed Hg, eventually precipitating HgS . The distribution of Hg between the two sites may reflect the efficiency and, therefore, the intensity of aqueous alteration on meteorite parent asteroids. More work is needed to determine the usefulness of Hg chemistry in tracking such parent-body alteration processes.

5.5. Se Release Profiles and Possible NAA Interferences

Kumar et al. (2001) suggested that measurements of Hg in meteorites by NAA may have failed to consider the interference between ^{75}Se and ^{203}Hg . ^{75}Se forms from ^{74}Se and ^{203}Hg from ^{202}Hg during neutron irradiation. ^{75}Se emits a γ -ray peak at 279.5 keV that overlaps with the ^{203}Hg peak at 279.2 keV. Failure to consider this interference could produce an apparent enrichment in ^{202}Hg , possibly resulting in the wide variability in reported isotopic ratios. To evaluate the contribution of Se to the Hg isotopic ratios measured by NAA, we simultaneously monitored the thermal release of ^{82}Se , ^{196}Hg , and ^{198}Hg .

The release profiles of ^{82}Se from both Allende and Murchison are different (Fig. 10). Allende releases large amounts of Se above $\sim 700^\circ\text{C}$ and the peak Se release occurs above 900°C . There is a small peak in the Se release profile from Allende at 400°C . Murchison, on the other hand, releases most of its Se at 410°C and also has a smaller peak at $\sim 700^\circ\text{C}$. In both meteorites, the amount of ^{82}Se released at 400°C is greater than or equal to the amount of ^{196}Hg released at this temperature.

The ionization potentials of Se and Hg are similar. Thus, the ratio of the count rates in ICP-MS is roughly equivalent to the elemental ratio. We calculated the corresponding ^{74}Se abundance using a $^{74}\text{Se}/^{82}\text{Se}$ ratio of 0.102 (Rossman and Taylor 1998) and the ^{202}Hg abundance using the $^{198}\text{Hg}/^{202}\text{Hg}$ ratio from the rightmost column of Table 3. The relative amounts of ^{75}Se and ^{203}Hg that would be produced in a thermal neutron flux were calculated using the corresponding σ values (52 and 4.9 barns, respectively). The contribution of ^{75}Se to the observed $\delta^{196}\text{Hg}$ values was determined by adding the amount of ^{75}Se produced during neutron irradiation to the ^{203}Hg abundance, taking into account the branching ratios at 279 keV for the two isotopes (25.2 and 81.5%). Based on this analysis we determine that failure to correct for a Se interference during NAA analyses of Hg isotope ratios could produce an apparent anomaly in $\delta^{196}\text{Hg}$ of -20% at 420°C from Allende and -4% at 435°C from Murchison. At higher temperatures, the largest apparent anomalies that could result from this interference are -130% at 863°C from Allende and -27% at 754°C from Murchison. However, many of the anomalies were reported from lower temperature fractions, where the contribution of ^{75}Se to the observed $\delta^{196}\text{Hg}$ value would be less than 1%. In addition, such an interference could not produce the positive anomalies reported for these meteorites. Finally, at least some of the studies of Hg isotopic ratios in meteorites using NAA corrected for this interference (G. W. Reed, personal communication; August, 2000). Thus, while an interference with Se may have been responsible for some of the anomalies reported in Allende or Murchison, it does not appear to explain all of them.

5.6. Implications for Hg Cosmochemistry

Elemental abundances (when normalized to Si and the composition of CI chondrites) in primitive meteorites correlate with the temperature at which they are predicted to condense from the solar nebula (Wasson and Kallemeyn, 1988). A recent study of Hg chemistry in the solar nebula suggests that it condensed between 300 and 525 K (Lauretta et al., 1999). Since this is lower than the condensation temperature of Si, CM and CV chondrites should be depleted in Hg, relative to CI chondrites. The abundance of Hg that we measured in Murchison results in a nearly solar Hg/Si ratio in this meteorite, suggesting a condensation temperature of $\sim 1200\text{ K}$, which is much higher than the value predicted by Lauretta et al. (1999). However, the lowest value reported for the abundance of Hg in Murchison (48 ppb) results in a condensation temperature of 178 K. This range of values suggests that the heterogeneity of Hg in Murchison is too wide for its abundance to accurately constrain the condensation temperature of Hg. The Hg abundance that we measured in Allende suggests that the Hg/Si ratio in this meteorite is $\sim 10\%$ of the solar value, yielding a condensation temperature of $\sim 375\text{ K}$, which is consistent with the calculations of Lauretta et al. (1999).

Several models have been put forward to explain the Hg isotopic anomalies measured by NAA in chondritic meteorites. These include a) production of the anomalous Hg by spallation reactions with elements having masses greater than 200, b) burn-up of ^{196}Hg by exposure to an intense thermalized neutron flux in the envelope of a supernova, and c) a multi-stage

distillation process in a circumstellar cloud (Jovanovic and Reed, 1976a). Distillation during evaporation and condensation in the solar nebula would produce mass-dependent isotopic fractionation, which was not observed in our experiments. The first two mechanisms would produce mass-independent Hg isotopic anomalies. However, these processes would alter several of the isotopic ratios, not just $^{200}\text{Hg}/^{202}\text{Hg}$. Further work is needed to determine if either of these mechanisms are responsible for the small Hg isotopic variation that we observed in Murchison.

6. SUMMARY

The bulk abundance and isotopic composition of Hg was determined in the carbonaceous chondrites Murchison (CM) and Allende (CV). Allende contains 30 ± 1.5 ng/g Hg and Murchison contains 294 ± 15 ng/g Hg. The isotopic compositions of both meteorites are identical to terrestrial values within the precision of our measurements (0.2–0.5‰) with one exception. The $^{200}\text{Hg}/^{202}\text{Hg}$ ratio appears to be slightly depleted in Murchison. The precision of these results places an upper limit on the abundance and isotopic composition of an anomalous Hg component. A hypothetical Hg fraction with $\delta^{196}\text{Hg} > 100\text{‰}$ in Allende could only comprise 0.4% of the total Hg.

Thermal release experiments reveal a distinct difference in Hg chemistry between Allende and Murchison. Allende releases Hg at two temperatures (225 and 344°C), whereas Murchison has only one peak (343°C). In both meteorites, the peak around 340°C correlates with a peak in the S thermal release patterns. This result suggests that HgS is a major Hg-bearing phase in both meteorites. The low-temperature release from Allende appears to reflect desorption of Hg from internal surfaces. The structure and position of the low-temperature peak is similar to that from basalt, suggesting that the Hg may be adsorbed on silicate grains. The release pattern for Allende is different from that of a sample of this meteorite that was heated and then exposed to Hg vapor for one hour, suggesting that contamination is not a factor. The distinct difference between the Hg chemistry of a CM versus a CV chondrite suggests that Hg chemistry may be a useful indicator of low-temperature processes on meteorite parent bodies and should be investigated further.

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Associate editor: U. Ott

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