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CHEMICAL FRACTIONATIONS IN METEORITES - IV.

ABUNDANCES OF FOURTEEN ~~TRACE~~ ELEMENTS IN L-CHONDRITES; IMPLICATIONS FOR COSMOTHERMOMETRY

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CHEMICAL FRACTIONATIONS IN METEORITES - IV. ABUNDANCES OF FOURTEEN TRACE ELEMENTS IN L-CHONDRITES; IMPLICATIONS FOR COSMOTHERMOMETRY

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Abstract A neutron activation procedure for Ag, Au, Bi, Br, Cd, Cs, Cu, Ga, In, Pd, Rb, Te, Tl, and Zn has been developed and applied to 12 chondrites (11 L3 to L6 and 1 LL3-4). Correlations between abundance and petrologic type were found for Bi, Br, Cs, In, and Tl. These elements correlated with each other and with primordial Ar and Xe. The observed inter-element correlations generally agree with calculated curves for trace element condensation from the solar nebula. It appears that most fractionations of volatile elements, including noble gases, arose in the solar nebula rather than in meteorite parent bodies. A few elements (Cs, Br, and Tl) are enriched in L3,4 chondrites above their cosmic abundance. Either these elements became enriched in the solar nebula toward the end of accretion, or they were volatilized in the meteorite parent bodies.

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Accretion temperatures of L-chondrites were estimated from Bi and Tl contents. Petrologic types 3 to 6 seem to have accreted in a 100-degree interval centered on 530^{+80}_{-60}°K. The error quoted reflects the uncertainties in total nebular pressure ($10^{-4\pm2}$ atm) and solubility of Bi and Tl in nickel-iron.

An equation for the fraction of In condensed ($\equiv \alpha_{In}$) was derived from the In data, using the accretion temperatures inferred from Bi and Tl contents: $\log[\alpha_{In}/(1-\alpha_{In})] = (8850\pm710)/T-(18.82\pm1.40)$.

INTRODUCTION

All volatile elements are fractionated in chondrites. The basic fractionation pattern has been explained by a two-component model (Anders, 1964, 1968; Larimer and Anders, 1967; Wood, 1963): chondrites being a mixture of a high-temperature fraction that has lost its volatiles (= chondrules, metal grains) and a low-temperature fraction that has retained them (= matrix). One feature that has remained controversial, however, is the strong depletion, by factors of 10^{-1} to 10^{-3}, of the most volatile group of elements in ordinary and enstatite chondrites. The depletion increases with volatility, reaching its peak for Pb, Bi, Tl, and In. It also increases with degree of recrystallization, rising steadily from petrologic type 3 to type 6 (Van Schmus and Wood, 1967; Tandon and Wasson, 1968; Dodd, 1969; Laul et al., 1970a,b).

Three explanations have been proposed for the abundance pattern of the "strongly depleted" elements.

1. Metamorphism. All ordinary chondrites initially had the composition and mineralogy of petrologic type 3. The depletion pattern was established during metamorphism, the most intensely metamorphosed meteorites losing the greatest proportion of volatiles (Wood, 1967; Dodd, 1969).

2. Condensation: 2 components. The pattern was established during accretion from the solar nebula. In a regime of falling temperatures, elements would condense in succession on the fine-

grained dust (= matrix). Meteorites last to accrete would therefore be richest in volatiles (Larimer and Anders, 1967; Anders, 1968). Being situated in the outermost layers of the parent body, they would also be least metamorphosed. Thus a strict, yet non-causal correlation between trace element depletion and metamorphism would result.

3. Condensation: 3 components. Abundances were established during accretion, as in the two-component model. However, strongly depleted elements were brought in by a third component, strongly enriched in all volatile elements (Tandon and Wasson, 1968).

Many of the arguments for and against these models have been based on inter-element correlations. Yet there exist surprisingly few data that can be used for such correlations. Most of the available data were measured on different samples in different laboratories. Apart from normal sampling problems owing to the coarse grain size of troilite and metal (Keil, 1962), there is the risk of sample mixups, compositional variations in single stones (particularly brecciated ones), and inter-laboratory errors.

In the course of developing a neutron activation procedure for trace elements expected to be depleted in lunar surface rocks, we had an opportunity to analyze a number of chondrites. Our procedure included the following 16 elements of which all but the last five are volatile: Ag, Bi, Br, Cd, Cs, Cu, Ga, In, Te, Tl, Zn, Au, Pd, Rb, Co, and Ir. Results for the first 14 are reported in this paper.

Originally, we had hoped to measure 3 representatives of each petrologic type from each of the 3 classes of ordinary chondrites. However, the arrival of the Apollo 11 samples terminated our project after only 11 L- and 1 LL-chondrite had been measured.

EXPERIMENTAL

Samples

Selection. In retrospect, our choice of samples was not especially fortunate. We selected 11 L-chondrites, more or less evenly divided among petrologic types L3 to L6. Nine of these had previously been analyzed for In and Ga by Tandon and Wasson (1968). Of the remaining two, Fukutomi had originally been classified as L5 by Van Schmus and Wood (1967) on the basis of an old and rather thick petrographic section. However, when our trace element data suggested an affinity to L4's, W. R. Van Schmus (private comm.) reexamined two new thin sections of this meteorite, and found it to possess all the characteristics of an L4, including basaltic glass. This left us with but four L5's and L6's.

Six of our meteorites (Bruderheim, Farmington, Homestead, Barratta, Goodland, and Modoc) had suffered some post-formational alteration. The first 5 were shocked, and the last three were finds. (Modoc is listed as an observed fall, but our sample was not recovered until several decades after the fall, and was noticeably weathered.) Our sample of Goodland, in addition to being shocked and a find, had once been subjected to "sulfur printing", involving

contact with acid-soaked photographic paper. It had very high Ag and Br contents, apparently reflecting contamination from the photographic emulsion. Thus more than half the meteorites used in our study were atypical in some respect.

Preparation. Samples were crushed to $\leq$2 mm size in an agate mortar, after removal of any fusion crust with a diamond burr. Each separately prepared sample is designated with a letter of the alphabet. Any aliquots derived from these samples are identified by consecutive numbers. Sample sizes ranged from 0.1 to 0.6 g. They were sealed in quartz vials, 3 to 5 mm I.D.

Monitors. Duplicate group monitors, containing 1-5 μg each of up to 6 elements, were prepared by weighing ~0.05-0.1 ml stock solution into quartz ampoules. In order to minimize self-shadowing, 1-50 mg Specpure MgO was added before evaporating the solution to dryness. The multi-element stock solutions were generally prepared by dissolving Specpure elements or compounds in HNO_3 or aqua regia, and diluting to volume with 1M HNO_3. For Bi and Pd, we used high-purity metals from the collection of the James Franck Institute, University of Chicago. The Br monitor consisted of a solution of $NaBrO_3$ in water.

The Zn values calculated on the basis of the original monitors seemed too low by about a factor of five. Specific activities of the monitors were reproducible but unreasonably high, suggesting an extraneous source of Zn. We suspect that zinc contamination was

present in one of the other elements included in the same group monitor solution: Cu, Bi, or Pd. When the Zn concentration in the stock solution was increased 200-fold, both the Zn results and monitor specific activities fell into line. The results were then recalculated on the basis of the Cs monitors.

Activity ratios of monitors were intercompared from run to run, in order to spot any anomalous values. On several occasions, particularly during the early runs, a few monitors were too low by a factor of up to two. Such values (which presumably were due to incomplete recovery from the irradiation vial) were corrected upward on the basis of the remaining monitors.

Irradiations. The first 2 sets of samples were irradiated at the Argonne CP-5 reactor, at a thermal neutron flux of 2×10^{13} cm^{-2} sec^{-1} for 10-17 days. The remaining irradiations were carried out at Oak Ridge, at 2×10^{14} n cm^{-2} sec^{-1} for 4-5 days. The ratio of fast-to-total neutron flux was ~100 in both reactors. Inasmuch as some of the shorter-lived activities (12.9-hr-Cu^{64}, 14.1-hr-Ga^{72}) had largely decayed during the 5-day cooling period required for radiological safety, the samples from the first two runs were re-irradiated for 30 minutes at 6×10^{13} n cm^{-2} sec^{-1}.

CHEMICAL PROCEDURE

Preliminary separations. The following procedure includes steps for Co and Ir which were measured in lunar samples but not in chondrites. The elements were divided more or less evenly

between the two senior authors by means of a group separation: R. R. Keays being responsible for Ag, Au, Bi, Cd, Cu, Ir, Pd, Tl, and Zn, while R. Ganapathy was responsible for Br, Cs, Ga, In, Rb, and Te.

The irradiated sample was fused in a zirconium crucible with 5 g NaOH + 5 g Na_2O_2 in the presence of 20-50 mg carrier for each element. After 10-30 min, when the sample had dissolved, the melt was allowed to cool and dissolved in about 50 ml water. The crucible was washed with H_2O and dilute H_2SO_4, and the washings were combined with the strongly basic melt solution. After boiling the solution briefly to destroy any residual peroxide, Ag, Au, Bi, Cd, Co, Cu, In, Tl, Ir, and Zn were precipitated by adding 5 ml 10% Na_2S and heating for 5 minutes. Ths sulfide-hydroxide precipitate was separated by centrifuging, washed twice with 20 ml 3M NaOH, and dissolved in aqua regia. The solution was evaporated twice to near dryness with HCl, and the residue taken up in 0.5M HCl containing Ce(IV) [to keep Ir in the (IV) state for the subsequent ion-exchange step].

The solution was centrifuged to remove AgCl and SiO_2, and was filtered onto a Dowex-1X8, 100 to 200 mesh column (1x10 cm). In, Co, and Cu passed through the column under these conditions. Residual amounts of these elements were eluted with 25 ml 0.75M HCl containing Ce(IV). Any remaining elutable impurities were removed by washing the column with 50 ml 2M HCl containing Fe and Co hold-backs, and 25 ml pure 2M HCl.

Zn was then eluted with 150 ml 0.1M HCl, Cd with 100 ml 0.002M HCl, Bi with 125 ml 1M H_2SO_4 containing 0.1% Ce(IV), and Tl with 125 ml 1M H_2SO_4 saturated with SO_2. Gold, which had been reduced to the metal by SO_2, was reoxidized to Au(III) by 15 ml aqua regia. (The aqua regia effluent contained some Ir and was therefore later combined with the main Ir fraction.) Au(III) was eluted with 150 ml of a solution 0.1M in HCl and 0.1M in thiourea. Finally, the major part of the Ir, having been reduced to Ir(III) by thiourea, was eluted with 200 ml 6M HCl. This solution was combined with the aqua regia fraction, above.

Principal contaminants. The most tenacious contaminants were Co, Ir, and Cr. The last two were present in nearly all fractions derived from the sulfide-hydroxide precipitate, Ir showing up most prominently with Bi and Tl, and Cr, with Cd and Zn. Cobalt tended to concentrate in the Cu fraction. Some difficulty was experienced in getting Ir and Cr holdback carriers to exchange with the active species.

Zinc. Holdbacks for Fe, Co, Cr, and Sc were added to the solution from the ion exchange step. The volume was reduced to 50 ml, the acidity adjusted to 2M HCl, $TiCl_3$ added to reduce Fe and Cr to +2 states, and the solution passed through another Dowex-1X8 column for final purification of Zn. $ZnHg(SCN)_4$ was then precipitated according to Hillebrand et al. (1953).

Cadmium. The Cd-fraction from the first ion exchange step was purified on another column, as for Zn. $CdNH_4PO_4 \cdot H_2O$ was precipitated from the effluent.

Bismuth. Holdbacks for Cr, Ir, Fe, and Co were added to the Bi-fraction. Bi_2S_3 was precipitated after appropriate adjustment of volume and pH. After dissolving the precipitate in aqua regia, 40 ml 4M HNO_3, 3 ml conc. HCl, and 10 mg Ir holdback carrier were added, and the solution heated to 100°C for 3-16 hrs to promote exchange of Ir (Kimberlin et al., 1968). The volume was reduced to near dryness, and the residue taken up in 25 ml 0.1M HCl. The solution was buffered with 2 g ammonium citrate, neutralized, complexed with 2 g KCN, and brought to pH 10. Bi was then extracted into $CHCl_3$ containing 0.15% dithizone.

The organic extract was washed carefully with a citrate buffer of pH 9.4, and the Bi back-extracted into 10% H_2SO_4. Excess dithizone was removed by several $CHCl_3$ extractions. Bismuth was then precipitated with NH_3, dissolved in 0.5M HNO_3, and precipitated as $BiPO_4$ with $(NH_4)_2HPO_4$.

Thallium. The Tl fraction was reduced to 50 ml and the acidity adjusted to 0.4 N. TlI was precipitated with KI, dissolved in aqua regia, and equilibrated with Ir holdback as described under Bi. The solution was allowed to go to dryness toward the end of the heating period. Nitrates were destroyed by heating with HBr, and Tl(I) oxidized to Tl(III) with 1M HBr + Br_2. Excess Br_2 was driven off as far as possible, Tl was extracted into diethyl ether, and brought back into an aqueous medium by evaporating the ether phase over 20 ml very dilute H_2SO_4. Organic matter was destroyed

by evaporation with aqua regia, excess HNO_3 was removed, and the residue was taken up in 0.1N H_2SO_4 saturated with SO_2. After two $La(OH)_3$ scavenges, TlI was precipitated from buffered acetate medium.

Gold. The Au-fraction was evaporated to dryness and the residue heated cautiously with 20 ml 6M HNO_3 to destroy thiourea. After reaction ceased, Ir holdback and conc. HCl were added. The solution was heated for 20 min to promote exchange. Nitrate ion was then destroyed by several evaporations with HCl. The final residue, heated to near dryness, was taken up in 30 ml 2M HCl. Metallic gold was precipitated with 0.5 g hydroquinone and purified by reprecipitation.

Iridium. The combined Ir fractions were taken to dryness. Any remaining NO_3^- was destroyed by two evaporations with HCl. Holdbacks for Fe, Cr, Co, and Sc were added; the pH was adjusted to 1.5 and the solution boiled for 5 min with 10 ml 10% $NaNO_2$. Base metals were then precipitated with $NaNO_2$ at pH 10 (Gilchrist, 1943). After destroying nitrite with HCl, iridium sulfide was precipitated from basic solution with Na_2S. The sulfide was dissolved and counted in a well crystal. The chemical yield was determined by re-activation, using an aliquot of the carrier solution as a monitor.

Silver. The AgCl + SiO_2 precipitate was washed with 1M HCl and dissolved in conc. NH_4OH. The solution was scavenged twice with $La(OH)_3$, and acidified with HNO_3. AgCl was then reprecipitated

with HCl. This cycle was repeated once. The precipitate was dissolved in NH_4OH; Cr, Co, and Fe holdbacks were added, and a mixture of sulfides and hydroxides was precipitated with Na_2S. The precipitate was washed with 2M HCl to redissolve Cr, Co, and Fe, and finally dissolved in HNO_3. After two scavenges with $Fe(OH)_3$ and $La(OH)_3$, AgCl was precipitated for counting.

Copper. The acidity of the Cu-Co-In fraction was raised to 2M HCl, CuS was precipitated, washed, and dissolved in 3M HNO_3. Holdbacks for As, Co, and Cr were added, and Cu was reprecipitated from 2M HCl after removal of NO_3^-. The cycle was repeated twice; then CuS was dissolved and the solution scavenged once with $Fe(OH)_3$ and twice with $La(OH)_3$. Finally, CuSCN was precipitated from 0.5M HCl in the presence of $NaHSO_3$.

Indium. The Co, In-containing supernatant from the CuS precipitation was boiled to expel H_2S. Fe^{3+} holdback was added, and the acidity adjusted to 10M HCl. The solution, typically 50 ml, was passed through a 1 cm x 60 cm ion exchange column of Dowex-1X8, 100-200 mesh. In was eluted with 400 ml 10M HCl, and Co, with 50 ml 5M HCl. The volume of the In-fraction was reduced to 5-10 ml, La carrier was added, and the pH was adjusted with NH_3 to 6.3-6.8 (brom-cresol purple endpoint). The mixed In-La hydroxide precipitate was centrifuged, and the co-precipitation with $La(OH)_3$ repeated on the supernatant after acidification and addition of more La carrier. The precipitate was taken up in

30 ml 4.5M HBr, and In was extracted into three 30-ml portions of iso-propyl ether. After three washings with 4.5M HBr, In was back-extracted into 3 portions of 6M HCl. Any trace of iron present remained in the organic phase. The aqueous phase was evaporated to 5-10 ml, and the whole extraction cycle was repeated. The final HCl extract was reduced to 1-2 ml volume and counted in a well crystal. The chemical yield was determined gravimetrically after counting, via the 8-hydroxyquinolate.

Cobalt. The volume of the cobalt fraction was reduced to 5-10 ml. $Co(OH)_2$ was precipitated with 10M KOH and dissolved in 6M acetic acid. After diluting the solution to about 3M, $K_3Co(NO_2)_6 \cdot H_2O$ was precipitated by standard gravimetric methods.

Gallium. Sufficient $Cd(NO_3)_2$ was added to the supernatant from the initial sulfide-hydroxide precipitation to remove all traces of $S^=$ as CdS. The pH of the solution was adjusted to 6.0 with H_2SO_4, to precipitate $Ga(OH)_3$ and $SiO_2 \cdot aq$. This left Rb, Cs, Br, and I in the supernatant. $Ga(OH)_3$ was dissolved in 6M HCl in the presence of Fe(III) holdback, and the solution was heated to promote exchange. $TiCl_3$ was added to reduce Fe(III) to Fe(II), and Ga was extracted twice into iso-propyl ether. Ga was back-extracted into water by 3 successive extractions, and precipitated as the 8-hydroxyquinolate.

If As was present as a radiochemical impurity, the extraction of Ga was repeated in the presence of As(V) holdback, without reduction by $TiCl_3$.

Rubidium and Cesium. The supernatant from the $Ga(OH)_3$ precipitation was acidified to 0.1N with H_2SO_4. Rb and Cs were precipitated with sodium tetraphenylboron. The precipitate was dissolved in 7.5M HNO_3 and heated with $HClO_4$ and HNO_3 to destroy organic matter. The solution was counted on a Ge(Li) detector. The chemical yield was determined by adding a known amount of Cs^{137} prior to fusion, and counting its 662-KeV γ-ray. Inasmuch as no perceptible fractionation of Rb and Cs occurs in this precipitation (H. Hamaguchi, private comm.; also Tomura et al., 1968) the chemical yield for Cs was assumed to apply to Rb as well.

Tellurium. Te was determined via the 8-day-I^{131} daughter of Te^{131}. The filtrate from the Rb + Cs precipitation was acidified with H_2SO_4, and IO_3^-, BrO_3^- were reduced to I^-, Br^- by sulfur dioxide. Nitrite was added to oxidize I^- to I_2, which was then extracted into CCl_4. The iodine was back-extracted into very dilute $NaHSO_3$, reoxidized with $NaNO_2$ in the presence of bromide holdback, and the cycle repeated. The aqueous extract was acidified with dil. HNO_3 and boiled to oxidize SO_2. Iodine was precipitated as AgI.

Bromine. A slight excess of $KMnO_4$ was added to the aqueous layer from the first iodine extraction, to oxidize Br^- to Br_2. It was purified by two cycles of extraction into CCl_4, and finally precipitated as AgBr.

Chemical yields. Typically, the following percentage yields were obtained. Ag 50-80; Au 40-50; Bi 40-50; Br 40-50; Cd 40-60;

Co 50-70; Cu 50-70; Ga 50-70; I 40-50; In 40-50; Ir 2-10; Rb + Cs 50-70; Tl 40-60; Zn 60-80.

Counting. The majority of nuclides were counted on single-channel γ-spectrometers with NaI crystals. In (and Ir in achondrites and lunar samples) were counted as solutions on a well crystal. Co, Rb, and Cs were measured on a Ge(Li) detector.

Bi, Tl, and Pd were counted on a low-level β-γ coincidence spectrometer (β^- background 0.25 cpm, β-γ background 0.01 to 0.03 cpm). The β-count was used for the actual activity measurement, and the β-γ coincidence count, as a check on radiochemical purity. Most neutron-induced activities, including the known major contaminant Ir^{192}, had γ's in coincidence with β's, and could therefore be detected with greatest sensitivity in the β-γ counting mode.

Radiochemical purity. Decay curves were generally followed for several days to months, except for Zn and the nuclides counted on a Ge(Li) detector. Whenever possible, γ-ray spectra were taken on a multi-channel analyzer, and compared with those of monitors. As the procedure was refined and modified, contamination problems became less severe. The following statements on radiochemical purity are representative of all runs on which data are reported.

Ag, Pd. No contaminants were observed in the Ag spectra. Decay was followed in order to resolve 7.5-day-Ag^{111} from 260-day-Ag^{110m}. Usually the resolution was based on the 658-KeV γ-peak alone, but sometimes β-counting data were also used. Apparent half-lives of Ag^{111} ranged from 6.5 to 8.0 days.

Au. Neither the decay curves nor the γ-ray spectra showed any evidence of contamination.

Bi. Iridium-192 and chromium-51 were sometimes present at the 0.01-0.1 cpm level, but could be resolved from the 5-day-Bi^{210} activity.

Br, I. No contaminants were seen after only a single extraction. The decontamination factor appeared to be $>5\times10^4$ for the first cycle. No extraneous activities were ever seen in any of the samples.

Cd. Some samples still contained Cr^{51}, visible in the decay curves though generally not in the initial γ-spectra. Although this contaminant could have been eliminated by recycling, it was tolerated so as not to delay separation of Bi and other short-lived activities.

Cs, Rb. Principal contaminants were Fe^{59}, Co^{60}, and Sc^{46}. The amounts were small, and were readily resolved on a Ge(Li) detector. We therefore did not attempt to remove these impurities, although it would have been an easy matter.

Cu. Except for small amounts of Co^{60} in some samples of low Cu content, no contaminants were present.

Ga. After one extraction, the sample still contained a few percent Fe^{59}. It was removed by a second extraction, to a total decontamination factor of $>10^6$.

In. The samples appeared to be radiochemically pure. The principal activities in the meteorite, Fe^{59} and Co^{60}, were present at levels below 1 dpm, implying a decontamination factor $>2\times10^9$.

Tl. The majority of samples decayed with the 3 8-yr half-life of Tl^{204} 1 to $1\frac{1}{2}$ yr after isolation. A few of the samples of very low initial activity decayed somewhat more rapidly, indicating slight contamination with Ir^{192}. Purity was always checked by γ- and β-γ coincidence counting.

Zn. Radiochemical purity was checked by γ-spectrum alone. This was deemed sufficient as the energy of the Zn^{65} photopeak is higher than that of either Cr^{51} or Ir^{192}. Fe^{59} and Co^{60}, two potential contaminants emitting high-energy γ-rays, were never detected in the samples.

RESULTS

Reliability of data

Earlier measurements on the same meteorites are given in Table 1. Our own results are shown in Table 2. Elements are arranged in order of increasing depletion in ordinary chondrites (Larimer and Anders, 1967), which also corresponds to the order of increasing volatility. The reliability of the data can be judged from three lines of evidence.

Geochemical standards. In the course of this work we analyzed duplicate samples of Columbia River Basalt BCR-1. The data, reported in Keays et al. (1970), agree well with accepted values (Flanagan, 1969).

Replicates. Because our procedure was still in the developmental stage, we ran from 2 to 6 replicates for each meteorite. Some of

these analyses were only partial, owing to contamination or monitor problems. The internal agreement is well within the usual range for 0.1 to 0.6 g samples of chondrites. Differences between aliquots are at least in part due to the coarse grain size, 1-2 mm.

Comparison with other work. Most of our data agree well with earlier measurements, where such are available. Exceptions are Bi and Tl, where differences by a factor of 2 or greater are found. The largest discrepancies are Bi and Tl in Barratta (both higher by a factor of 5 than the values of Laul et al., 1970a,b) and Tl in Modoc (our 4 values are higher than 6 of the 7 values by Reed et al., 1960 and Ehmann and Huizenga, 1959). On checking our results, we found no obvious source of error. The Tl samples from Modoc decayed with the half-life of Tl^{204}, and the specific activities of the monitors agreed with expected values. Contamination can never be completely excluded, but the identity of our and Laul's Bi/Tl ratio in Barratta, and the constancy of our Tl values in Modoc places rather improbable constraints on any contaminant. As far as Modoc is concerned, we suspect that our sample became contaminated in the years between fall and recovery. The Barratta discrepancy may be due to a sample mixup.

Gross trends

Our results are plotted in Fig. 1, again in the order of increasing depletion in (equilibrated) ordinary chondrites. We have also included data for Ar, Xe, and C, taken from the literature.

The data were normalized to Cl chondrites, with a correction for the higher Si content of L-chondrites (18.69 vs. 10.63%). For Ar and Xe, the silicate phase of the Orgueil Cl chondrite was used for normalization: $Ar^{36} = 217 \times 10^{-8}$ and $Xe^{132} = 3.3 \times 10^{-8}$ ccSTP/g (Jeffery and Anders, 1970).

Following Tandon and Wasson (1968) we have used mnemonic symbols for the four petrologic types, the type number being indicated by the number of sides of the polygon. (Type 5 is represented by a five-pointed star instead of a pentagon, for reasons that are wholly morphological, not political.) As a guide to the eye, tie lines were drawn through the points for the most depleted meteorite, Bruderheim.

For purposes of nomenclature, it will be useful to classify these elements into groups (Larimer and Anders, 1967, 1970).

Undepleted elements (Rb, Pd, Au, and others not included here) are relatively non-volatile, and are present in nearly their cosmic or (Cl chondrite) abundances. (The slight deficiency of siderophile Pd and Au probably reflects loss of metal phase from L-chondrites during metal-silicate fractionation.)

Normally depleted elements (Cu, Ga, and others not included here) occur at a mean abundance of ~0.25. This would be expected if the chondrites consisted of 25% matrix containing Cu,Ga in cosmic proportions, and 75% Cu-,Ga-free chondrules-plus-metal.

Strongly depleted elements (the remaining 12) have mean

abundances of less than ~0.15. In terms of the two-component model, they are underabundant even in the matrix.

Several trends may be noted.

(1) The dispersion of the points is correlated with depletion. The first five, "undepleted" and "normally depleted", elements cluster tightly. The next twelve, "strongly depleted", elements show increasing dispersion, which on the whole parallels the degree of depletion.

(2) Among the strongly depleted elements, the first three (Ag, Te, Zn) as well as Cd show no correlation between depletion and petrologic type. The remaining five (Cs, Br, Bi, Tl, and In) show a fairly good correlation.

(3) Some meteorites contain greater amounts of strongly depleted elements than expected from the two-component model. If the chondrites contained 25% matrix of cosmic composition, and 75% volatile-free chondrules, no volatile element should be present in an abundance greater than 25% of cosmic. Yet 7 of the Cs points, 3 of the Br points, and one of the Tl points lie above this limit.

DISCUSSION

Inter-element correlations: Predicted

The three models mentioned in the Introduction predict somewhat different correlations for strongly depleted elements. Let us review these predictions before examining the data in detail.

Three-component model. The simplest correlation is expected

for the Tandon-Wasson model. If the strongly depleted elements were brought in wholly via a trace-element-rich fraction of fixed composition, then these elements should occur in fixed proportions. On logarithmic, two-element correlation plots, the points should always fall along 45° lines.

Two-component model. A wider range of possibilities is expected for the two-component model of Larimer and Anders. Condensation of a pure element E follows the relation:

$$\log(1 - \alpha) = -A'/T + B' \qquad (1)$$

where α is the fraction condensed, T is the absolute temperature, and A' and B' are constants. [$A' = \Delta H_v/2.303R$, and $B' = b - \log P_t - \log(2E/H)$, where ΔH_v is the heat of vaporization, R is the gas constant, b is the constant of integration in the Clausius-Clapeyron equation, P_t is the total pressure in the solar nebula, and E,H are the atomic abundances of element E and H in the nebula.] In the more common case where a trace element forms a solid solution with a major phase M, the condensation equation is:

$$\log\frac{\alpha}{1-\alpha} = \frac{A}{T} - B \qquad (2)$$

The symbols have the same meaning as before, except that A now contains an additional term ΔH_s, the heat of solution of E in M: $A = \Delta H/2.303R = (\Delta H_v - \Delta H_s)/2.303R$. B, in turn, equals $b - \log P_t - \log(2M/H)$.

In the range of incomplete condensation, where $1-\alpha \approx 1$, linear correlations may be expected between two volatile elements:

$$\log\alpha_1/\alpha_2 \approx \frac{1}{2.303RT}(\Delta H_1 - \Delta H_2) - (B_1 - B_2)$$

Because of the temperature dependence of the first term, a $\log\alpha_1$ vs. $\log\alpha_2$ plot will generally have a slope greater or less than 1, depending on whether $\Delta H_1 - \Delta H_2$ is positive or negative. Only in the special case where $\Delta H_1 = \Delta H_2$ will a slope of 1 be observed, and even then only in the range where the approximation $1-\alpha \approx 1$ holds for both elements. In actual practice, though, differences between ΔH_1 and ΔH_2 are generally small, and the correspondingly small deviations of the slope from unity may be hidden by scatter of the data.

Several theoretical condensation curves for Bi and Tl are shown in Fig. 2a. They are based on the revised condensation diagram of Larimer and Anders (J. W. Larimer, to be published; Anders, 1968). The constants for Bi and Tl are: A' = 9701, 8750; B' = 20.07, 20.19; A = 6100, 5690; B = 13.25, 13.20. They are based on $P_t = 10^{-4}$ atm; Cameron's (1968) cosmic abundances, and estimated heats of solution in nickel-iron of 16.5±1.5 and 14±1 kcal/mole for Bi and Tl. Additional curves are shown for $P_t = 10^{-2}$ and 10^{-6} atm and for the upper and lower limits of ΔH_s.

Owing to the limited solubility of these metals in nickel-iron, the condensation proceeds first according to eq. (1) and then, when the nickel-iron has become saturated, according to eq. (2).

The resulting curves are fairly similar in shape. At low degrees of condensation, they are virtually linear. Slopes are close to 45°, owing to the similarity in ΔH. As condensation progresses, the curves flatten abruptly and then turn upward, asymptotically approaching a 90° slope. The inflection points correspond to the solubility limit of the less volatile trace element in nickel-iron. These limits are only approximately known, being based on extrapolations from other metallurgical systems (O. J. Kleppa, private comm.; J. W. Larimer, to be published). A change in ΔH_s corresponding to the estimated limits of error (±1 kcal/mole for Tl and ±1.5 kcal/mole for Bi) shifts the inflection points considerably (dashed lines in Fig. 2a). But the general shape of the curves is certainly typical of partially immiscible systems.

We can construct at least approximate correlation curves for noble gases. Complete solubility data are unavailable, but comparison of measurements on enstatite melts at 1773°K (Kirsten, 1968), and magnetite at 500°K (M. S. Lancet, to be published) suggests a $\Delta H_{Ar} \approx 15$ to 20 kcal/mole. We have therefore calculated three condensation curves for Ar^{36}, with ΔH = 15, 20, and 25 kcal/mole (A = 3279, 4372, and 5464; B = 15.68, 18.05, and 20.42). The constant B was chosen so as to give an Ar^{36} solubility of 15×10^{-8} ccSTP/g at 496°K. (This is about a factor of 100 higher than the value predicted from Lancet's data for magnetite at P_t =

10^{-4} atm. In order to eliminate this discrepancy, one may postulate higher pressures or a higher argon solubility in silicate than in magnetite.)

Correlation curves of Ar^{36} with Bi are shown in Fig. 2b. Again, the correlation is virtually linear over a surprisingly large part of its range. The curves turn sharply to the right after the solubility limit of Bi is reached. But as Fig. 2a shows, the position of the inflection point depends strongly on P_t and ΔH_s. At higher pressures or higher solubilities, the inflection point would disappear altogether. In that case, the curves would turn gently upward, as condensation of Bi approached completion. A small non-linearity of this sort might, however, go unnoticed in a plot of actual experimental data.

Unfortunately, it is not possible to calculate condensation curves for the remaining elements of interest: Cs, Br, In, Cd, and C. The first two are probably lithophile, but their chemical states, host phases, and solid-solution relationships are not known. Indium is largely siderophile, but its condensation is retarded by the existence of a volatile sulfide, In_2S (Larimer, 1967). In order to assess its effects one must know the partial pressure of H_2S, which depends not only on the completeness of the reaction $Fe + H_2S \xrightarrow{<680\,^\circ K} FeS + H_2$, but also on the distribution of sulfur among various gaseous species (H_2S, CS_2, COS, S_2, S_8, etc.).

Cadmium and carbon are still more difficult to handle. Cd is chalcophile and hence condenses during FeS formation, but the

uncertainties in solid-solution relationships and abundance of gaseous sulfur species preclude quantitative treatment. Carbon presumably condensed by catalytic (Fischer-Tropsch) and radiation-induced formation of high-molecular weight organic compounds (Urey, 1953; Studier et al., 1968). The degree of condensation should hence depend on surface area as well as temperature.

Metamorphic Model. This model offers few a priori predictions. If equilibrium is reached and conditions are otherwise equal, metamorphic loss and fractional condensation will produce exactly the same fractionation pattern, because volatilization and condensation must lead to the same equilibrium state. But for conditions to be equal, the gas phase in the meteorite parent body must be of cosmic composition, and at a pressure equal to that in the nebula ($\sim 10^{-4}$ atm). The system cannot have been open to the nebula, for otherwise FeS would have disappeared from all regions of the body that were heated to more than 680°K during metamorphism. If the system was closed, with the gas phase comprising an "internal atmosphere" filling the pores (DuFresne and Anders, 1962), the composition of the gas would soon depart from cosmic, owing to reaction of the gas with solid phases. Such reactions can produce major shifts in gas phase composition, because the ratio gas/solid in the body would be only about 10^{-13} that in the nebula. Consequently it is virtually certain that the condensation curves of Fig. 2 cannot be applied to metamorphic fractionations.

Dodd (1969) has made a few general statements about fractionations expected during metamorphism. Losses are expected during recrystallization or chemical change of a phase. If two volatile elements are concentrated entirely in such a phase, they will be lost in a 1:1 ratio; if they are distributed over several phases, they will be lost in variable ratios. Some part of the volatiles may be retained in their original proportions in highly retentive sites.

Inter-element correlations: observed

Tl, In, Bi. Plots of Tl and In vs. Bi are shown in Figs. 3a,b. Data by Laul et al. (1970a,b) are also included, and are represented by open symbols. We have not plotted the data of Reed et al. (1960) and Ehmann and Huizenga (1959). About half of their values fit the trend of our data; the others scatter rather widely, with Tl tending to be high. The two L-chondrites measured by them, Holbrook and Modoc, were showers whose recovery extended over several decades. It is possible that some samples used by these authors had suffered some post-fall alteration, like our Modoc sample. Dashed lines in the upper right represent "maximum" abundances expected in a chondrite containing 25% matrix of essentially cosmic composition (= same trace element/silicon ratio as cosmic matter; Cameron, 1968). The diagonal lines are predicted Tl-Bi condensation curves, for those combinations of P_t and ΔH_s giving best agreement with the data.

We can probably disregard our two Modoc points, as our weathered

sample seems to have been contaminated with Tl. The remaining points by and large are consistent with all three models. Except for Krymka, they fit the 45° line of the Tandon-Wasson model about as well as the condensation curves of the Larimer-Anders model. The 45° line intercepts the "maximum" abundance for Bi (dashed vertical line) at 15 ppb Tl; only one-quarter the maximum for Tl. In terms of the Tandon-Wasson model this would imply that their fraction C was four-fold enriched in Bi relative to Tl.

The region from 2 to 50 ppb Bi may eventually provide a crucial test between the two models. The Tandon-Wasson model predicts a correlation with a slope of 1, while the Larimer-Anders model predicts some degree of flattening, with Tl rising less rapidly than Bi. The present data show a hint of such flattening, but the scarcity of points in the crucial interval 10-50 ppb precludes any firm conclusions. The present data are consistent with all 4 curves shown, if we bear in mind that the cosmic abundances of Bi and Tl to which these curves are tied are uncertain by perhaps 50%.

Krymka is a puzzling case on any model. It contains more Tl than Bi, contrary to the volatility trend. Similar Tl excesses were noted by Laul et al. (1970a,b) in the least equilibrated Type 3's of other chemical groups, but were interpreted by them as Bi deficiencies. We shall return to this point later on.

The In-Bi plot (Fig. 3b) fails to show the 45° trend required

by the 3-component model. Whether the data are consistent with the 2-componen. model cannot be determined in the absence of a calculated condensation curve. The trend for L3's and L4's can be approximated rather well by a curve similar to the Tl-Bi curve in Fig. 2, but the steep drop between L4's and L5's requires some special mechanism. Perhaps this represents the effect of the volatile sulfide In_2S which is expected to retard condensation of In at higher temperatures, so long as sufficient S is present in the gas phase. Upon exhaustion of S by the reaction $Fe + H_2S \rightarrow FeS + H_2$, In would revert to more normal condensation behavior.

Ar, Xe, Bi, In. Correlation plots of the first three elements are shown in Figs. 4a,b. Some adjustments were made in the Ar^{36} data, to compensate for gas losses in heavily shocked meteorites. All cases with $Ar^{36}/Xe^{132} \leq 80$ (open symbols) were corrected upward to an Ar^{36}/Xe^{132} ratio of 150, an approximate average for unshocked meteorites. This procedure will result in an undercorrection in those cases where Xe^{132} was lost as well, but there is no reliable way to estimate such losses.

There is no single trend line that represents all the data. The 3-component model predicts a 45° line, which, however, misses Farmington, Homestead, Tennasilm, and Mezö-Madaras. Part of the discrepancy may be due to sampling, the noble-gas and trace element measurements having been done on different samples, from different museums. The 2-component model predicts a curve of more complex

shape (cf. Fig. 2b), the slope depending on ΔH for the noble gas, and the inflection point, on P_t and ΔH_s for the metal. Four such curves are shown in Figs. 4a,b. They, too, miss Farmington and Homestead, but pass rather close to Tennasilm and Mezö-Madaras and generally follow the flat trend of the data above 4 ppb Bi. The heavy curves, representing minimum solubility of Bi (ΔH_s = 18 kcal/mole), were shifted slightly to the right, in order to make their linear portions pass through the L6 points. This is tantamount to increasing the cosmic abundance of Bi by a factor of 1.75, but an equivalent shift could have been achieved by a slight increase in P_t or the solubility of Bi in nickel-iron. A Xe-In plot (Fig. 5) shows a slope decidedly less than 45°. This is also true for Tandon and Wasson's (1968) In data, which are represented by open symbols. Although Tandon and Wasson drew a 45° line through their data, the points deviate systematically from the line. All of the In points at the low end fall below the line, and most of the points at the high end, above. A slope of ~30° is indicated for both sets of data.

Br, Cs, Bi. Theoretical condensation curves are unavailable for the first two elements. The Bi-Br plot (Fig. 6a) has a slope steeper than 45°, and in fact somewhat resembles the condensation curves for elements with limited solubility in a major phase (Fig. 2). The Br values quickly approach the maximum, while Bi values keep rising. However, four of the bromine values lie above the

expected maximum abundance (dashed line). An even more striking overabundance is shown by Cs, where 14 of 24 points fall above the maximum (Fig. 6b). All of the meteorites in question are L3's and L4's.

Perhaps this overabundance implies that the chondrules of these meteorites had not lost their Cs, having formed at lower temperatures. However, a similar overabundance had previously been noted for several other highly volatile elements: Hg in most ordinary chondrites (Reed and Jovanovic, 1966; Ehmann, 1967) and In in the chondrites Chainpur (LL3; Schmitt and Smith, 1968), Hallingeberg (L3; Tandon and Wasson, 1968), and the dark portion of the gas-rich chondrite Leighton (H5; Rieder and Wänke, 1969).

Both the metamorphism and condensation models can account for it by appropriate ad hoc assumptions. According to the metamorphic model, these elements may have volatilized from the hot interior (source of L6's) and recondensed in the cool outer regions (source of L3,4's). Whether this explanation is satisfactory in detail remains to be seen. Temperatures for volatilization in a gas-tight asteroid differ from element to element (Larimer and Anders, 1967; Table 5). It is not clear that volatilization of this sort will in fact produce the observed enrichments, and no others. Somehow the excess Bi, Tl, In, and Cd distilled from the inner parts of the asteroid must be prevented from recondensing in the outer layers, to prevent a buildup of abundances above cosmic, while other

elements (Hg, Cs, Br) are allowed to recondense in order to achieve precisely such a buildup. This requires that Bi, Tl, In, and Cd were more volatile in the parent body than were Hg, Cs, and Br. Some shifts in the volatility sequence are in fact expected, because the lower H_2/H_2O and H_2/H_2S ratios (Mueller, 1964; Dodd, 1969) in the parent body will cause changes in geochemical character and hence mineralogical siting of some elements. But the changes are likely to be in the wrong direction. In, Bi, and Tl, which are siderophile in the solar nebula, are expected to be chalcophile in the parent body (cf. Fig. 3 of Larimer, 1967). [In fact, Fouché and Smales (1967a) have shown that In is chalcophile in equilibrated chondrites.] Since these elements are likely to be more soluble in troilite than in nickel-iron, their volatilities in the parent body ought to be reduced. Conversely, the ready leachability of Cs and Br (Gast, 1960; Smales et al., 1964; Reed and Allen, 1966) suggests that these elements are not located in major phases where their volatility would be reduced by solid-solution effects, but in a minor phase. Thus their volatilities should be enhanced relative to those of Bi, Tl, In; just the reverse of what is required by the metamorphic model.

According to the condensation model, the overabundances may have been established either in the nebula or in the parent body. As condensation and accretion progress, the gas phase becomes enriched in the volatile complement of both chondrules and matrix

of early-accreted meteorites. If only a small fraction (<30%) of the potentially accretable material actually accretes, the volatile complement of the accreted material will not greatly change the composition of the nebular gas. Solid grains equilibrating with this gas at falling temperatures thus will gradually approach cosmic composition, without ever appreciably exceeding it.

If a large fraction of the solid material accretes, the complementary gas will be increasingly enriched in volatiles as accretion progresses. For example, if accretion is 90% complete, the remaining gas will be 10-fold enriched in residual volatiles. This mechanism can, in principle, lead to enrichment of the most volatile elements, such as Hg and In. It is probably not suitable for Cs and Br, unless we are prepared to assume that these elements ranked between In and Hg in volatility.

It seems more likely that the overabundance of Cs and Br was established during metamorphism. In attempting to account for all fractionations by metamorphism, we found that the apparent volatility reversal of the Pb-Bi-Tl-In and Cs-Br groups made it impossible to enrich the latter in colder regions (L3,4) without enriching the former in somewhat warmer regions (L4,5). However, in the condensation model the burden of Pb-Bi-Tl-In fractionations is shifted from the parent body to the nebula. Hence temperatures in the parent body need only be high enough to fractionate Cs, Br, and Hg.

Larimer and Anders (1967) had previously inferred from the

absence of trace element enrichments that the accretion efficiency was low, as argued by Cameron (1962) on astrophysical grounds. However, now that a few instances of such enrichment have been found for In and Tl, this conclusion stands in some doubt, at least insofar as the final stages of accretion are concerned.

In-Cs; Bi-C. These two element pairs also correlate, though the second one only weakly so (Fig. 7a,b). The slope is greater than 1 in both cases, contrary to the three-component model. The interpretation is not straightforward, however. Cesium is over-abundant in L3's and L4's, as discussed above, while carbon fails to reach the expected maximum (1/4 the Cl abundance) even in the least equilibrated L3's. As noted earlier, the condensation behavior of carbon is likely to differ from that of other elements, being dependent on surface area as well as temperature.

Thermometry

Bi and Tl. We can calculate accretion temperatures for each meteorite, using the Bi and Tl contents from Table 2 and the condensation equations for these elements. The results are given in Table 3. These values apply to a total pressure of 10^{-4} atm. Temperatures at the upper and lower pressure limits estimated for the solar nebula (10^{-2} and 10^{-6} atm; Larimer and Anders, 1970), are some 80° higher and 60° lower.

On the whole, the Bi- and Tl-based temperatures agree rather well with each other, although the latter tend to be systematically

lower. It appears that the temperature span for accretion of ordinary chondrites was only ~100°K.

In. Indium is a potentially valuable cosmothermometer, because it is volatile, highly fractionated among meteorites, and readily measurable. Unfortunately, its condensation curve is not well-determined, owing to complexities in its chemistry. We can try to estimate this curve from our data, tying the In abundances to the Bi-Tl temperature scale.

It appears that the condensation of In followed eq. (2), the relation for solid-solution formation: $\log[\alpha/(1-\alpha)] = A/T - B$. A plot of $\log[\alpha/(1-\alpha)]$ vs. 1/T is essentially linear (Fig. 8). The equation of the line (excluding Tennasilm, which deviates strongly from the trend) is

$$\log[\alpha/(1-\alpha)] = (8850\pm710)/T - (18.82\pm1.40) \qquad (3)$$

With Tennasilm, the equation is:

$$\log[\alpha/(1-\alpha)] = (7500\pm760)/T - (16.24\pm1.53) \qquad (4)$$

Temperatures were obtained from Bi and Tl data (including literature values) and were averaged for each meteorite. In values were taken from Table 2 as well as Tandon and Wasson (1968) and Schmitt and Smith (1968).

Equation (3) can be used to estimate accretion temperatures from In contents. These temperatures are not truly independent,

as the In scale has been tied to the average of the Bi and Tl scales. However, the dispersion of the points about the line is great enough to suggest the possibility of independent trends. Indeed, some interesting differences show up in a histogram of Bi-, Tl-, and In-based accretion temperatures (Fig. 9). On the Bi and Tl scales, L5's are associated with L4's; on the In scale, they are associated with L6's. The separation between L3's and L4's is sharper on the In scale than on the Bi or Tl scales. This reflects the fact that the L4 chondrite Tennasilm has an In content normal for an L4, though its Bi and Tl contents are well in the L3 range.

These temperature estimates must be regarded as preliminary, being based on quite a small number of meteorites. With additional measurements, it should be possible to improve our estimates of P_t and ΔH_s, and thus obtain better concordancy among the different cosmothermometers. But even from the present data it seems safe to conclude that the L-chondrites accreted in a 100-degree interval centered on $\sim 510^{+80}_{-60}$ °K.

CONCLUSIONS

Of the three models mentioned in the Introduction, the 3-component model of Tandon and Wasson (1968) can probably be ruled out on the basis of the present data. Several of the correlation plots (e.g. Figs 3b, 5, and 6a) show slopes other than 45°, a trend inconsistent with this model.

The choice between the metamorphic and two-component condensation models is less clear-cut, mainly because there are so few specific predictions associated with the former model. Most of the observed trends agree well with correlations predicted from the condensation model. Several distinctive irregularities are present which probably require ad hoc explanations on any model.

There is no evidence for a genetic or compositional hiatus between L3's and L4-6's, as proposed by Dodd (1969). Although trace element ratios frequently vary between the two groups, this variation is of the order predicted by the condensation model. Aberrant points are no more common among L3's than among L4-6's. The present evidence is consistent with the view that these meteorites comprise a single genetic sequence.

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Table 1. Results by Other Authors

Meteorite	Class	Rb (ppm)	Pd (ppm)	Au (ppb)	Cu (ppm)	Ga (ppm)	Te (ppm)	Ag (ppb)	Zn (ppm)	Cs (ppb)	Br (ppb)	Bi (ppb)	Tl (ppb)	In (ppb)
Khohar	L3				112 n	6.0kk			157 n			31.0 u	2.0 v	14±1 gg
					93±14ee	5.9kk								21 kk
					105±6 ff									20 kk
Krymka	L3					5.0kk						17.0 u	50.7 v	8.2 kk
						5.0kk								8.7 kk
Mezö-Madaras				200bb	230 bb	6.2bb						123 u	3.6 v	13±1 gg
					97±5 ff	4.8kk								22 kk
						4.6kk								21 kk
Hamlet	LL3-4											6.5 u	1.1 v	
Barratta	L4	6.0 b			79.4 n	4.9kk			76.3n			1.58u	0.25v	8.2 kk
					63±15ee	4.9kk								11.2 kk
					56±9 ff	5.1kk								7.9 kk
					88±7 ff	4.9kk								11.9 kk
						4.6kk								9.4 kk
Tennasilm	L4				91±7 ff	5.5kk								5.4 kk
						5.5kk								4.9 kk
Farmington	L5			120bb	56.8 n	4.9bb			102 n					0.176 kk
					193 bb	5.1kk								0.171 kk
					62±10ee	6.0kk								
					92±10ff									
Homestead	L5	3.6 r		120bb	67.3 n	4.1bb	0.46nn		63.8n	56ii				0.186 kk
		2.8lii			210 bb	5.5kk				50jj				00.26 kk
		3.1 jj			185±50ee	5.7kk				70jj				
					114±20ff									

Table 1. (continued)

Meteorite	Class	Rb (ppm)	Pd (ppm)	Au (ppb)	Cu (ppm)	Ga (ppm)	Te (ppm)	Ag (ppb)	Zn (ppm)	Cs (ppb)	Br (ppb)	Bi (ppb)	Tl (ppb)	In (ppb)
Modoc	L6	3.45±0.06f	0.30e	91e	92±6 ff	4.7d		120dd		90±7f		3.0 c	4.2 c	0.5 d
		3.41 g	0.82p			5.4kk				84 ii		0.07c	0.24c	0.6 dd
		2.97 g	0.63p			5.4kk				80 jj		0.33c	0.41c	0.3±0.1gg
		2.95 g	0.73p									0.4 z	0.32z	0.24 kk
		3.0 jj										1.0 z	0.03z	0.152 kk
		3.15 kk										6.2 z		
Bruderheim	L6		600 e	170a	98 m	4.9e	0.46±0.03h	58k	56k		180 i	1.4 u	<0.06v	1.3 d
			1000 k	130a	109 m	5.5n	0.20±0.01h		50m		180 i			0.033 kk
			500 hh	190e	114 m	5.7ii	0.50±0.03h		52y		150 i			0.092 kk
					95±6 ff	5.1ii	0.76 k		50y		160 i			
					93±8 ff		0.67 x				109±19i			
					94±12ff		1.96 t				120±30i			
					99±8 ff		0.50±0.02aa				30 w			
											26 w			
											20 x			
											130±10aa			
											52±7 aa			
											197±10aa			
											240±30aa			
											225±10aa			
											970 mm			
											1560 mm			

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h. Goles and Anders (1962).

i. Goles et al. (1967).

j. Greenland (1965).

k. Greenland (1967).

m. Greenland and Goles (1965).

n. Greenland and Lovering (1965).

p. Hamaguchi et al. (1961).

r. Herzog and Pinson (1956).

t. Kiesl et al. (1970).

u. Laul et al. (1970a).

v. Laul et al. (1970b).

w. Lieberman and Ehmann (1967).

x. Merrihue (1966).

y. Nishimura and Sandell (1964).

z. Reed et al. (1960).

aa. Reed and Allen (1966).

bb. Schaudy et al. (1968).

cc. Schindewolf (1960).

dd. Schindewolf and Wahlgren (1960).

ee. Schmitt et al. (1970).

ff. Schmitt and Smith (1966).

gg. Schmitt and Smith (1968).

hh. Sen Gupta (1967).

ii. Smales et al. (1964).

jj. Smales et al. (1958).

kk. Tandon and Wasson (1968).

mm. Wyttenbach et al. (1965).

Table 2. Results

Meteorite*		Source, Number†	Class (PMD)‡	Rb (ppm)	Pd (ppm)	Au (ppb)	Cu (ppm)	Ga (ppm)	Te (ppm)	Ag (ppb)	Zn (ppm)	Cs (ppb)	Br (ppb)	Cd (ppb)	Bi (ppb)	Tl (ppb)	In (ppb)	Gas content§ 10^{-8} cc STP/g Ar^{36}_{p}	Xe^{132}_{p}
Khohar	A1	AS623.1	L3	2.3	0.44	118	67	-	0.14	94	56	384	4070	75	5.4	1.7	8.0	25b	16b
			(18)																
	A2			3.6	0.57	158	77	-	0.18	199	57	572	7070	63	6.0	2.4	11.2		
Krymka	A	CM	L3	-	0.46	148	99	-	-	151	-	-	-	-	-	112	-	49bc	29bc
			(45)																
	B			3.0	0.59	142	86	-	0.59	98	55	158	1810	25	10.7	65	13.1		
	C			4.2	0.60	155	93	-	0.61	95	75	207	2150	24	10.7	118	18.4		
Mezö-Madaras	A1	AS340.2	L3	2.6	0.70	259	93	5.0	0.97	1350	65	268	1880	34	80	6.6	25.5	34bc	21abc
			(21)																
	A2			2.4	0.66	285	94	4.5	0.78	861	68	210	1550	29	63	5.4	-		
Hamlet	A	AT	LL3,4	-	0.56	111	85	-	-	-	-	-	-	-	-	-	-	9.5e	9e
	B		(3.5)	-	0.55	174	83	-	-	-	-	-	-	-	-	-	-		
	C			3.1	0.43	-	72	4.9	0.41	177	51	110	1560	56	2.5	1.9	-		
	D			2.7	0.39	142	77	5.1	0.60	78	51	125	2130	106	3.6	2.4	-		
Barratta	A	F1463	L4	3.1	0.45	114	70	6.3	0.21	111	58	291	6000	35	8.3	0.84	8.1	5.6bch	14.2bcf
	B		(4.2)	3.0	0.38	158	67	6.0	0.21	96	57	298	7200	11	7.2	0.97	7.2		
	C			-	-	109	-	-	-	-	63	-	-	-	-	-	-		
Fukutomi	A1	F1491	L4	4.9	0.51	166	96	5.3	0.41	36	-	581	1940	31	3.1	0.60	5.4		
	A2			5.3	0.50	168	81	5.8	0.43	38	73	619	2140	30	4.1	0.92	5.8		
Goodland	A	AM425.14	L4	-	0.69	201	89	-	-	-	-	-	-	-	-	1.7	-	2.7b	10b
	B		(2.6)	2.4	-	170	71	6.4	0.69	-	66	214	1090	7.4	4.0	-	-		
	C			2.2	-	278	88	6.4	0.93	-	63	207	1360	8.6	2.1	0.56	-		
	D			-	-	398	-	-	-	-	70	-	-	-	-	-	-		
Tennasilm	A1	F1874	L4	3.1	0.47	285	76	5.0	0.43	57	159	340	1100	85	38	4.6	2.4	5.6i	7i
	A2			3.0	0.60	142	98	5.2	0.55	50	110	330	990	30	44‖	6.0	2.3		

Table 2. (continued)

Meteorite*	Source,† Number	Class (PMD)‡	Rb (ppm)	Pd (ppm)	Au (ppb)	Cu (ppm)	Ga (ppm)	Te (ppm)	Ag (ppb)	Zn (ppm)	Cs (ppb)	Br (ppb)	Cd (ppb)	Bi (ppb)	Tl (ppb)	In (ppb)	Gas Content§ 10^{-8} ccSTP/g Ar^{36}_{p}	Xe^{132}_{p}
Farmington A1	F345	L5	2.2	0.55	177	86	5.1	0.38	62	59	23	44	50	1.5	1.0	0.21	0.4h	0.88d
A2			2.2	0.56	166	82	5.7	0.38	73	58	23	43	54	-	0.77	0.22		
Homestead A1	F313	L5	1.4	0.61	152	100	-	0.43	149	57	55	940	103	3.0	1.1	0.28	1.2h	0.3i
A2			3.7	0.75	91	100	-	0.40	161	56	59	950	129	3.4	1.0	0.42		
Modoc A	AS81.3	L6	-	0.52	98	66	5.7	0.40	105	68	-	350	7.1	-	1.1	-	1.1h	-
B			-	0.56	121	80	6.9	0.47	116	-	-	700	15	-	1.4	-		
C			4.0	0.62	156	95	-	0.46	99	67	72	820	7.4	0.34	1.2	0.16		
D			2.9	0.59	155	75	-	0.46	108	52	53	860	10.8	0.23	1.3	0.33		
Bruderheim A	W B137	L6	2.4	0.69	173	75	6.5	0.56	49	-	4.0	92	-	≤0.4	-	-	0.7gh	1.1eg
B			3.1	0.72	180	79	6.6	0.58	75	-	4.8	128	-	≤0.2	0.11	-		
C			-	0.67	178	56	6.7	0.53	82	-	-	73	-	-	0.07	-		
D			-	0.64	177	57	6.5	0.49	50	-	-	110	-	-	0.10	-		
E1			2.7	0.58	158	61	-	0.47	53	56	10.3	120	15	0.14	0.06	0.09		
E2			2.5	0.78	207	113	-	0.57	63	53	13.1	115	18	0.47	0.05	0.06		

Notes to Table 2

*Letters refer to different chips from the same stone; numbers to coarsely crushed aliquots from the same chip.

†AM = American Meteorite Museum, Sedona, Arizona (Dr. H. H. Nininger)
AS = Arizona State University, Tempe, Arizona (Dr. C. B. Moore)
AT = Dr. A. Turkevich, University of Chicago, Chicago, Illinois
CM = Committee on Meteorites, Acad. of Sci. U.S.S.R. (Dr. E. L. Krinov)
F = Field Museum of Natural History (Dr. E. J. Olsen)
W.= Ward's Natural Science Establishment, Rochester, New York

‡PMD = percent mean deviation of iron content in olivine (Dodd et al., 1967)

§Gas contents were taken from the following references:

a. Eugster et al. (1968)
b. Heymann and Mazor (1968)
c. Marti (1967a)
d. Marti (1967b)
e. Marti et al. (1966)
f. Müller and Zähringer (1969)
g. Zähringer (1962)
h. Zähringer (1966)
i. Zähringer (1968)

‖Probably contaminated.

Table 3. Accretion Temperatures for L-Chondrites

($P_t = 10^{-4}$ atm)

Meteorite	Class	α^*_{Bi}	α^*_{Tl}	α^*_{In}	Temperature, °K Bi	Tl	In
Khohar A1	L3	0.137	0.0395	0.279	490	482	481
Khohar A2		0.133	0.0497	0.341	491	477	477
Krymka B		0.212	1.20	0.357	483	-	477
Krymka C		0.197	2.01	0.465	483	-	472
Mezö-Madaras A1	L3	1.47	0.112	0.646	-	462	464
Mezö-Madaras A2		1.22	0.0959	-	-	465	-
Hamlet C	LL3-4	0.0535	0.0368	-	508	483	-
Hamlet D		0.0707	0.0449	-	503	479	-
Barratta A	L4	0.154	0.0143	0.205	488	501	485
Barratta B		0.139	0.0172	0.190	490	497	486
Fukutomi A1	L4	0.0543	0.00977	0.131	508	508	492
Fukutomi A2	L4	0.0741	0.0154	0.144	502	499	490
Goodland B	L4	-	0.0298	-	-	487	-
Goodland C		0.0732	-	-	502	-	-
Goodland D		0.0341	0.0086	-	517	511	-
Tennasilm A1	L4	0.782	0.0869	0.0670	469	467	499
Tennasilm A2		0.783	0.0974	0.0560	469	465	503
Farmington A1	L5	0.0276	0.0180	0.00546	521	496	534
Farmington A2		-	0.0129	0.00550	-	503	534
Homestead A1	L5	0.0512	0.0175	0.00657	509	497	532
Homestead A2		0.0581	0.0164	0.00986	507	498	526

Table 3. (continued)

Meteorite	Class	α^*_{Bi}	α^*_{Tl}	α^*_{In}	Temperature, °K Bi	Tl	In
Modoc C	L6	0.0061	-	0.00395	553	-	540
Modoc D		0.0052	-	0.0103	556	-	526
Bruderheim A	L6	≤0.0070	-	-	≥550	-	-
Bruderheim B		≤0.0034	0.0017	-	≥566	545	-
Bruderheim C		-	0.0012	-	-	553	-
Bruderheim D		-	0.0018	-	-	544	-
Bruderheim E1		0.0039	0.0014	0.00345	563	549	541
Bruderheim E2		0.0071	0.0007	0.00125	549	566	556

*α = fraction condensed. From Cameron's (1968) cosmic abundances, 100% condensation of Bi, Tl, and In would result in abundances of 57, 62, and 42 ppb in an L-chondrite containing 25% matrix. In calculating α values, we attempted to estimate actual matrix contents of each sample from Ga and Cu contents; 100% matrix corresponding to 389 ppm Cu and 21.1 ppm Ga.

Figure Captions

Fig. 1. Trace element abundances in 11 L-chondrites. The first 5 elements ("undepleted" or "normally depleted") show little variation with petrologic type. The next 12 ("strongly depleted") show much larger variations, and often tend to decrease in abundance from L3 to L6. Data from Table 2 except carbon values, which are from Moore and Lewis (1967) and Otting and Zähringer (1967).

Fig. 2a. Theoretical condensation curves for Tl and Bi. At low degrees of condensation, the correlation curve has a slope near 1. Once the solubility limit of Bi in nickel-iron is reached, the curves change slope. Solubility limit and hence position of inflection point depends on total pressure and heat of solution ΔH_s. Solid curves are based on ΔH_s = 14 and 16.5 kcal/mole for Tl and Bi; upper dashed curve (= $\Delta H_{s,min}$) on 13 and 15 kcal/mole; lower dashed curve (= $\Delta H_{s,max}$) on 15 and 18 kcal/mole.

Fig. 2b. Estimated condensation curves for Ar and Bi. The three curves differ in the assumed value of ΔH.

Fig. 3a. Observed trend of Tl and Bi data is consistent with predictions of 3-component model (45° line, not plotted) or 2-component model (curves actually shown). Dashed lines in upper right correspond to expected maximum abundances (= full

cosmic complement) in a chondrite with 25% matrix. Tl in Krymka appears to be overabundant. Tl values for Modoc should be regarded as upper limits, owing to possible contamination. Open symbols: data of Laul et al. (1970a,b). Abbreviations: Ba = Barratta, Br = Bruderheim, Fa = Farmington, Fu = Fukutomi, Go = Goodland, Ha = Hamlet, Ho = Homestead, Kh = Khohar, Kr = Krymka, MM = Mezö-Madaras, Mo = Modoc, Te = Tennasilm.

Fig. 3b. In-Bi plot has slope steeper than 45°, contrary to prediction of 3-component model.

Fig. 4a,b. Ar and Xe data deviate somewhat from predictions of 3-component model (45° line) or 2-component model (solid curves). Ar^{36} values of meteorites with $Ar^{36}/Xe^{132} \leq 80$ (open symbols) were corrected upward to $Ar^{36}/Xe^{132} = 150$, to compensate for shock-induced gas losses. Numbers on condensation curves refer to ΔH_s for Bi and ΔH for Ar or Xe, both in kcal/mole. In order to improve the fit, curves with $\Delta H_s = 18$ were arbitrarily shifted to the right, corresponding to an increase in the cosmic abundance of Bi by a factor of 1.75. However, an equivalent improvement might have been achieved by changing ΔH_s to 17 or 17.5 kcal/mole.

Vertical bars give observed range of noble-gas contents. Data were taken from Zähringer's (1968) review.

Fig. 5. Xe-In plot has a slope less than 45°, contrary to predictions of 3-component model. Open symbols represent data of Tandon and Wasson (1968). PR = Peace River; Cy = Cynthiana; Bj = Bjurböle; He = Hedjaz; Ma = Manych; Bi = Bishunpur.

Fig. 6a. Br and Bi show a rough correlation. Some meteorites contain more than the maximum (dashed line) expected from cosmic abundances and a matrix content of 25%.

Fig. 6b. Br and Cs correlate with petrologic type and with each other, but Cs is overabundant in 14 of 24 meteorites. This may imply redistribution of Cs, Br in the meteorite parent body, or volatile-enrichment of the nebula in the terminal stages of accretion.

Fig. 7a. In-Cs correlation has a slope greater than 45°, contrary to the prediction of the 3-component model.

Fig. 7b. Bi and C show only a weak correlation. Carbon values from Moore and Lewis (1967) and Otting and Zähringer (1967).

Fig. 8. Indium abundance correlates with accretion temperature inferred from Bi and Tl. This suggests that In condensed according to the same relation as did other volatile metals: $\log\alpha/(1-\alpha) = A/T - B$, where α = fraction condensed. (Open symbols: data by Tandon and Wasson, 1968, and Schmitt and Smith, 1968.)

Fig. 9. Accretion temperatures of 11 L-chondrites from Bi, Tl, and In contents. Circles: data of Tandon and Wasson (1968) and Schmitt and Smith (1968). Meteorites are those listed in Table 3.

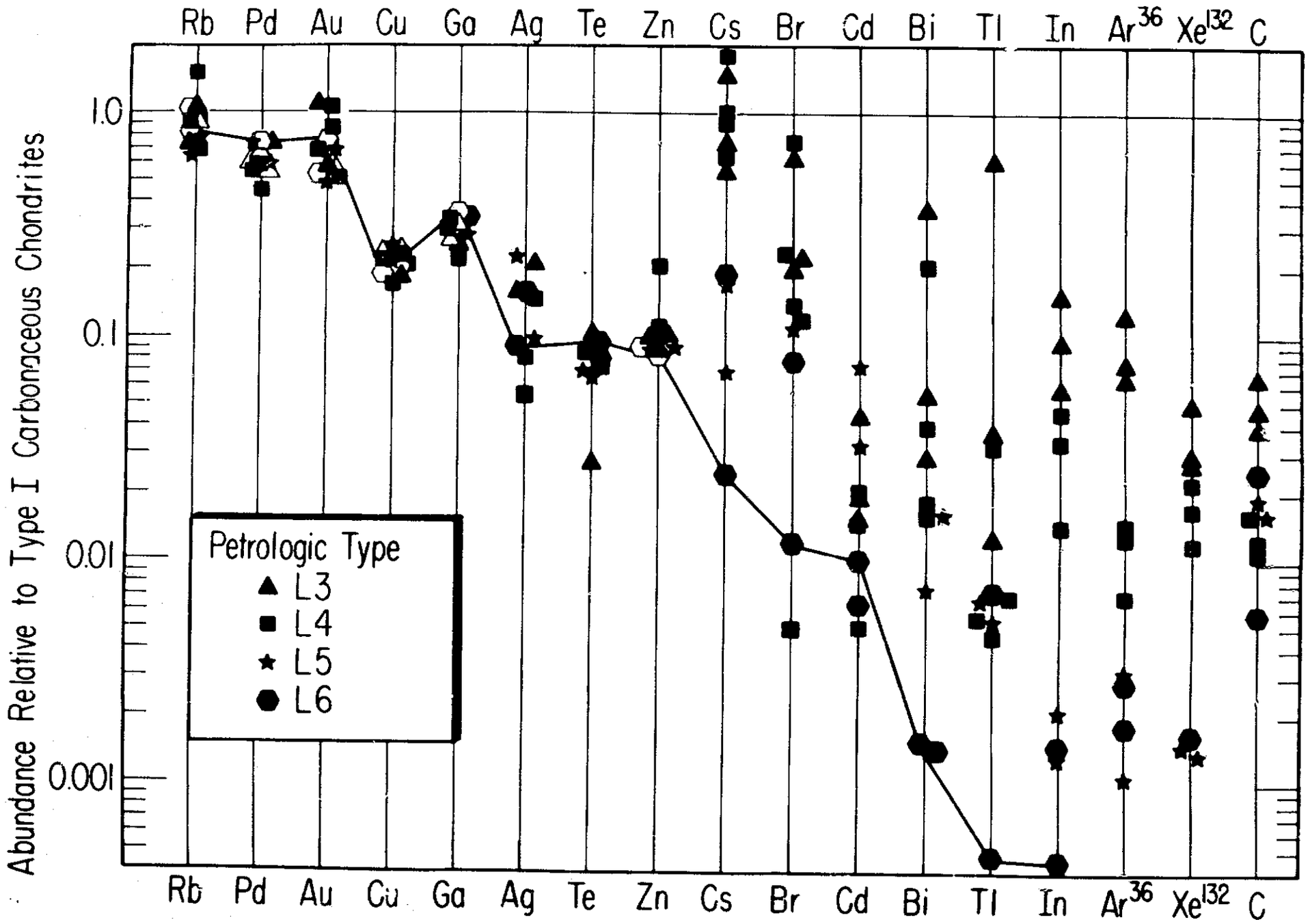

Rb Pd Au Cu Ga Ag Te Zn Cs Br Cd Bi Tl In Ar36 Xe132 C
1.0
0.1
0.01
0.001
Abundance Relative to Type I Carbonaceous Chondrites
Petrologic Type
L3
L4
L5
L6

Fig.1

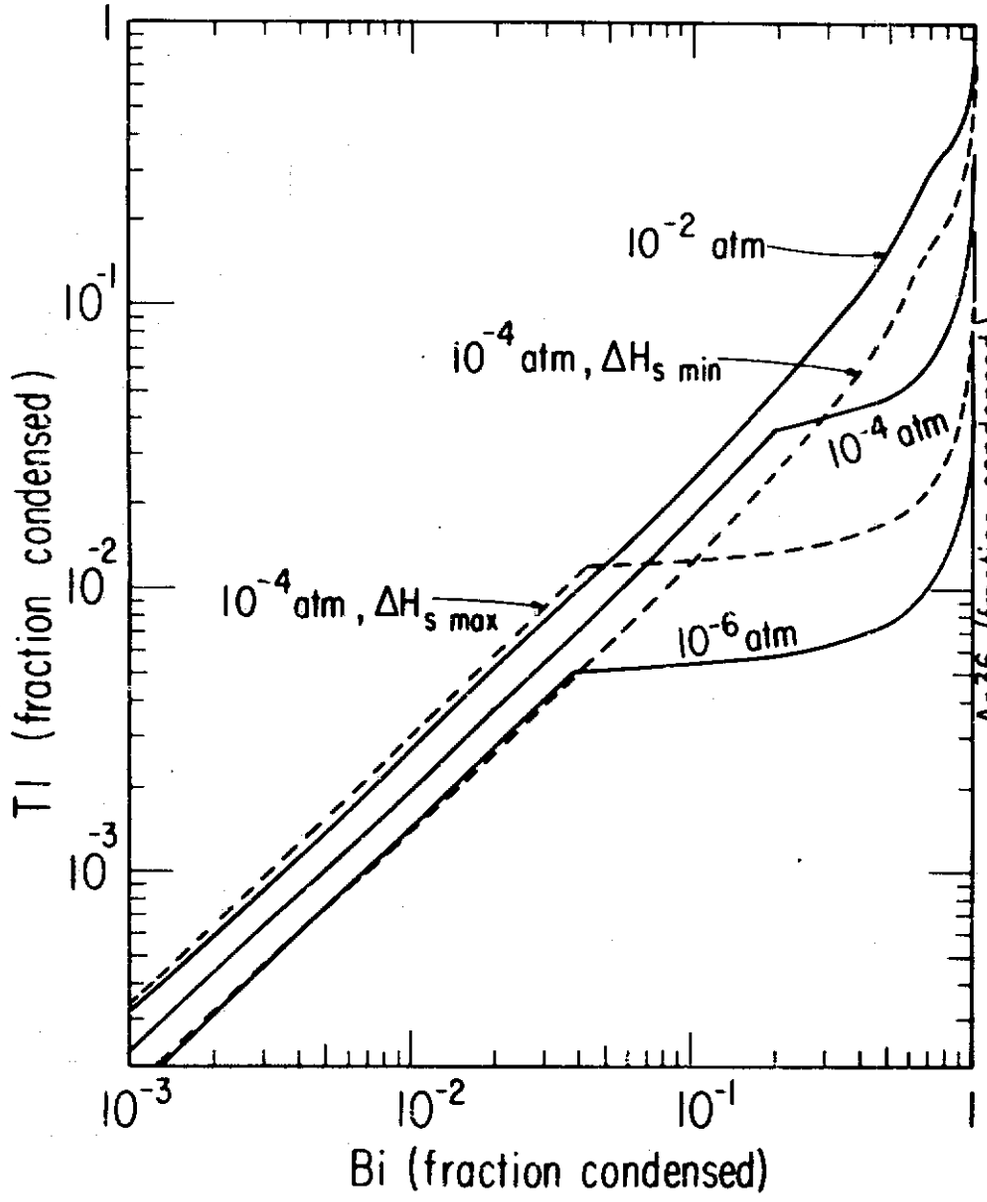

1
10^{-1}
10^{-2}
10^{-3}
TI (fraction condensed)
10^{-2} atm
10^{-4} atm, $\Delta H_{s\ min}$
10^{-4} atm
10^{-4} atm, $\Delta H_{s\ max}$
10^{-6} atm
10^{-3}
10^{-2}
10^{-1}
1
Bi (fraction condensed)

Fig.2(a)

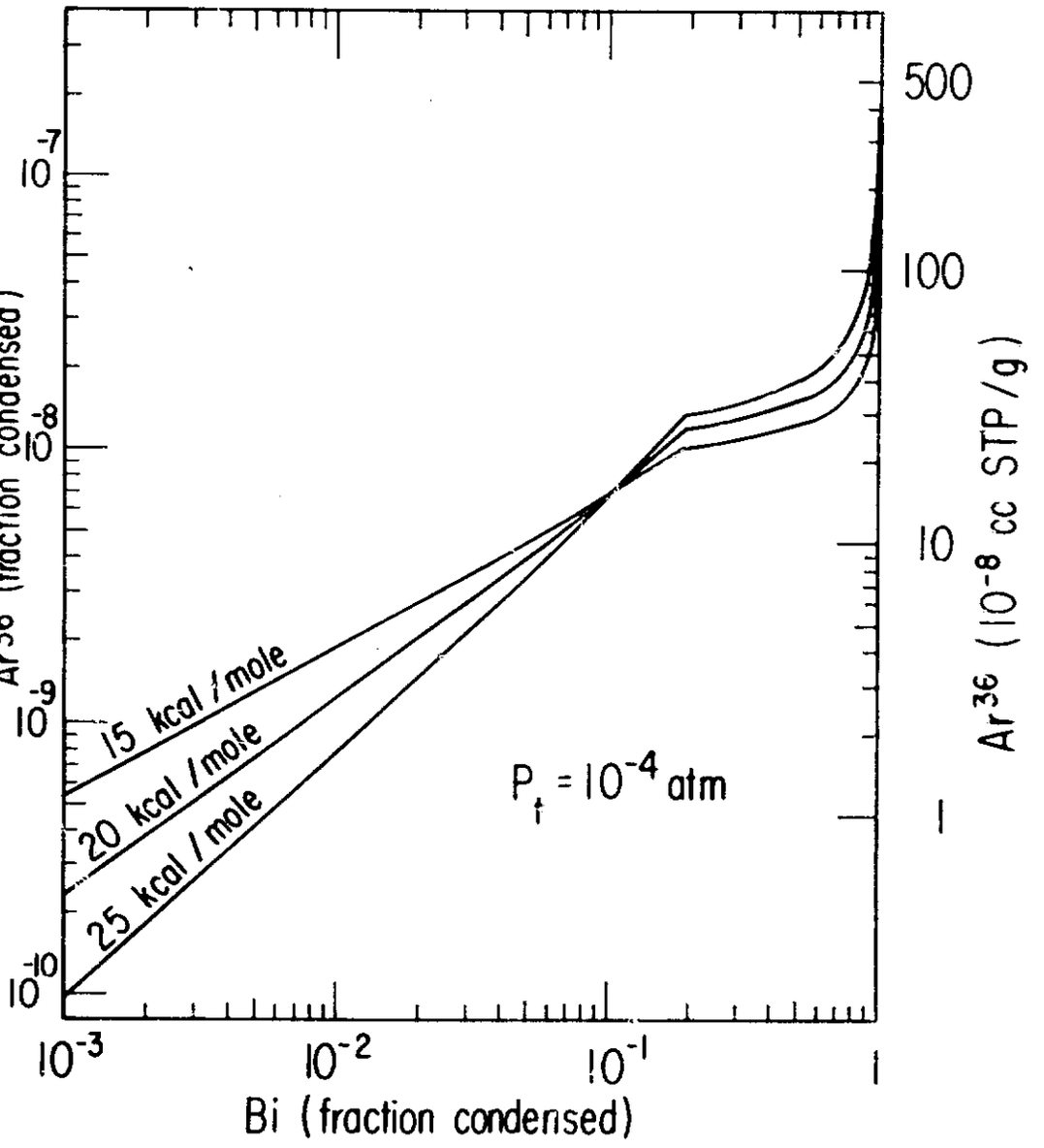

10^{-7}
10^{-8}
10^{-9}
10^{-10}
Ar^{36} (fraction condensed)
15 kcal / mole
20 kcal / mole
25 kcal / mole
$P_t = 10^{-4}$ atm
10^{-3}
10^{-2}
10^{-1}
1
Bi (fraction condensed)
500
100
10
1
Ar^{36} (10^{-8} cc STP/g)

Fig.2(b)

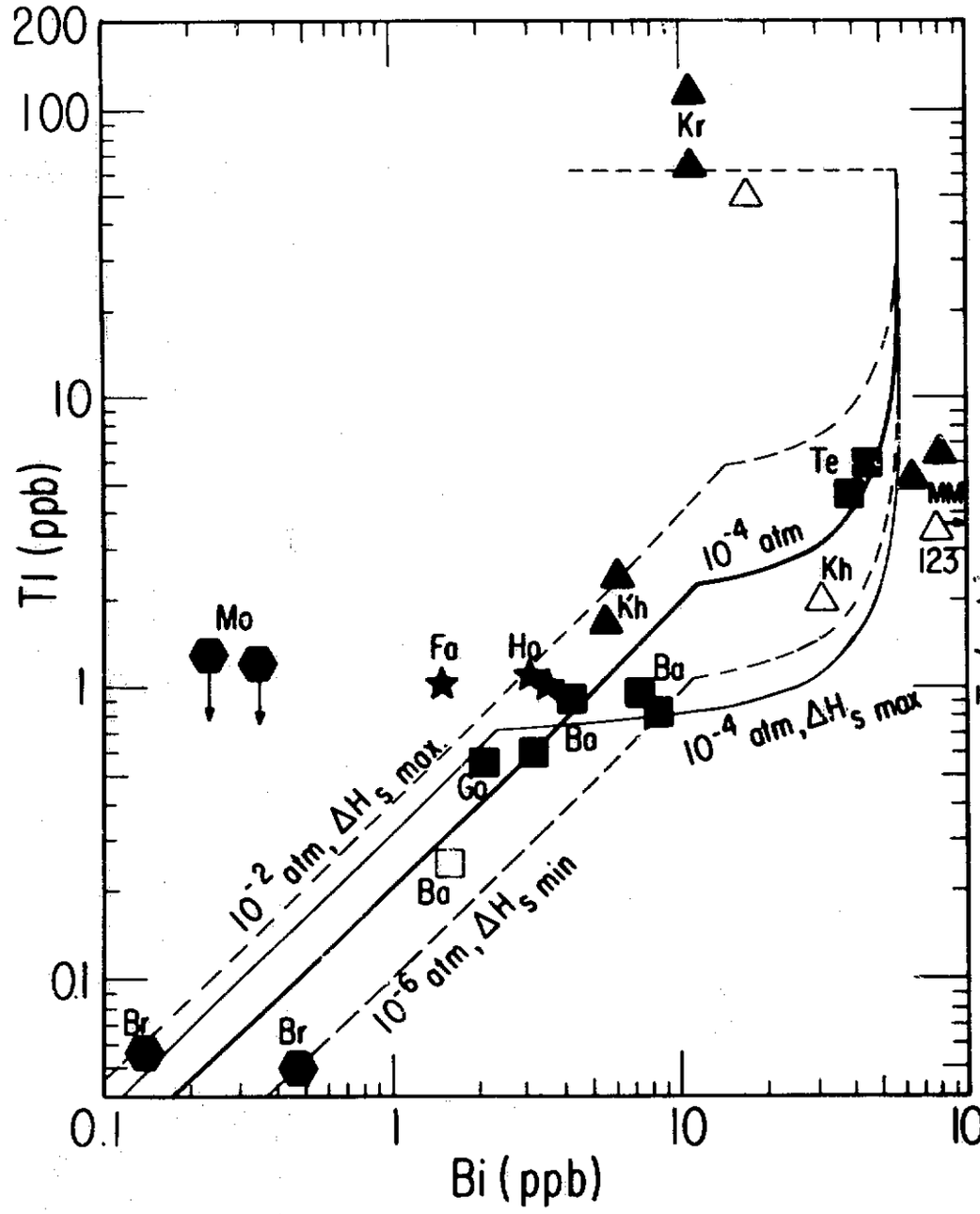

200
100
10
1
0.1
Tl (ppb)
Kr
Te
MM
123
Kh
Mo
Fa
Ho
Ba
Go
Br
10^{-4} atm
10^{-2} atm, ΔH_s max.
10^{-4} atm, ΔH_s max
10^{-6} atm, ΔH_s min
0.1
1
10
100
Bi (ppb)

Fig.3(a)

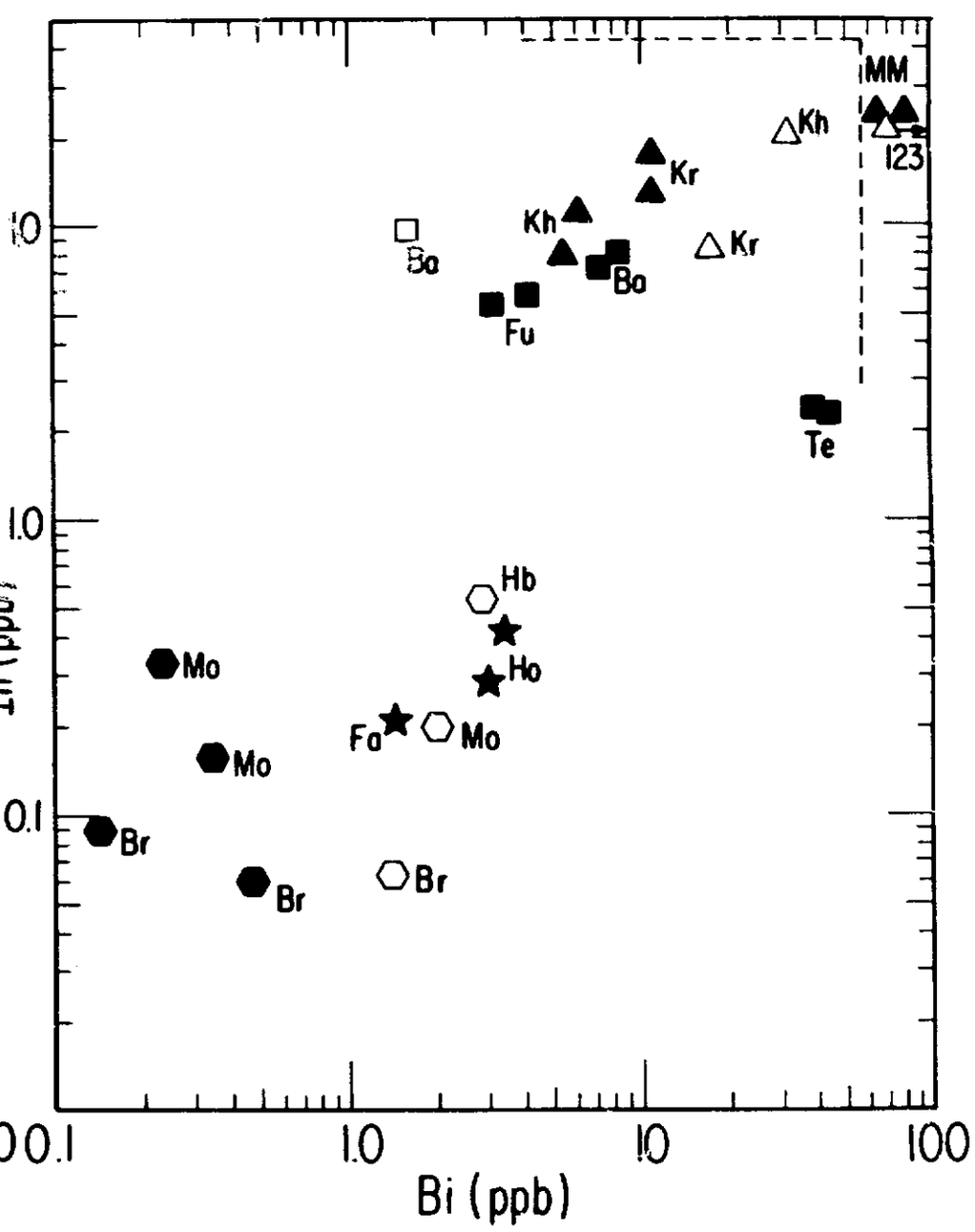

MM
Kh
123
Kr
Ba
Fu
Te
Hb
Ho
Mo
Fa
Br
10
1.0
0.1
In (ppb)
0.1
1.0
10
100
Bi (ppb)

Fig.3(b)

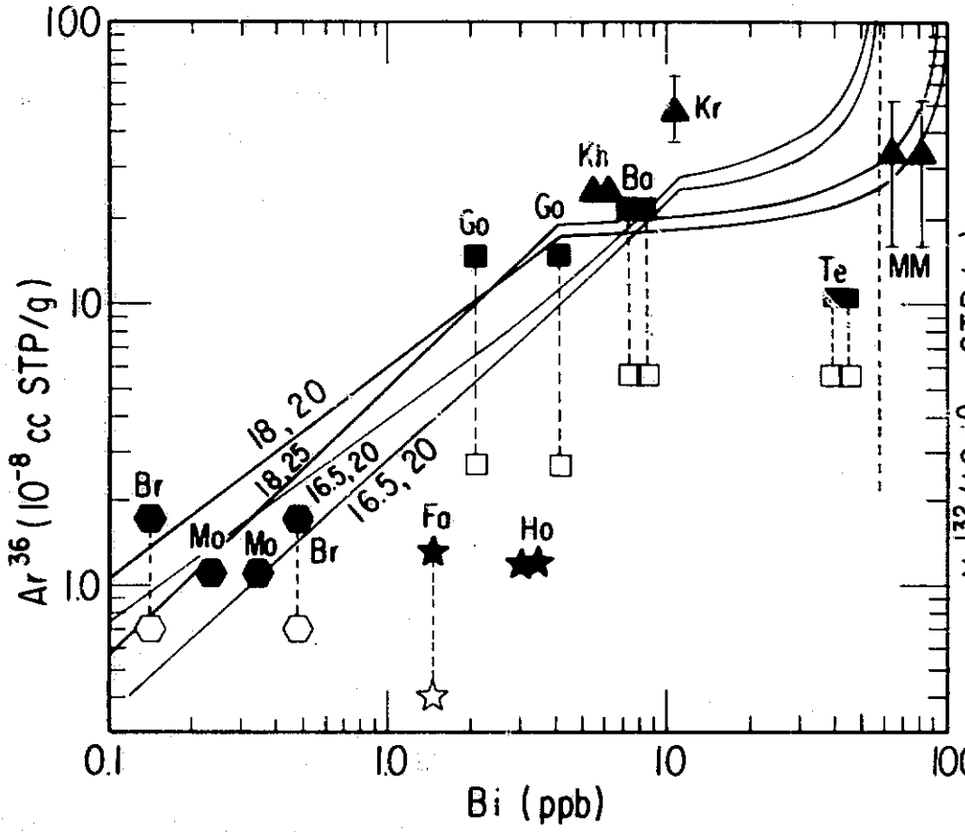

100
10
1.0
0.1
1.0
10
100
Bi (ppb)
Ar36 (10-8 cc STP/g)
Kr
Kh
Ba
Go
Go
Te
MM
Br
Mo
Mo
Br
Fa
Ho
18, 20
18, 25
16.5, 20
16.5, 20

Fig.4(a)

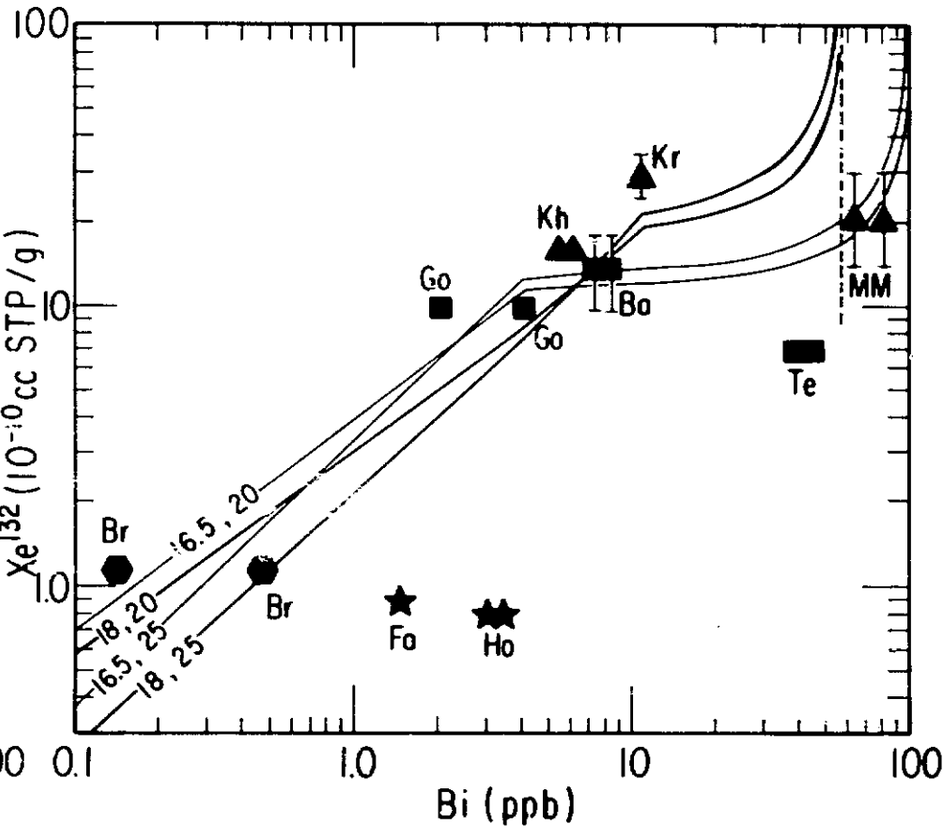

100
10
1.0
0.1
1.0
10
100
Bi (ppb)
Xe132 (10-10 cc STP/g)
Kr
Kh
Go
Ba
Go
Te
MM
Br
Br
Fa
Ho
16.5, 20
18, 20
16.5, 25
18, 25

Fig.4(b)

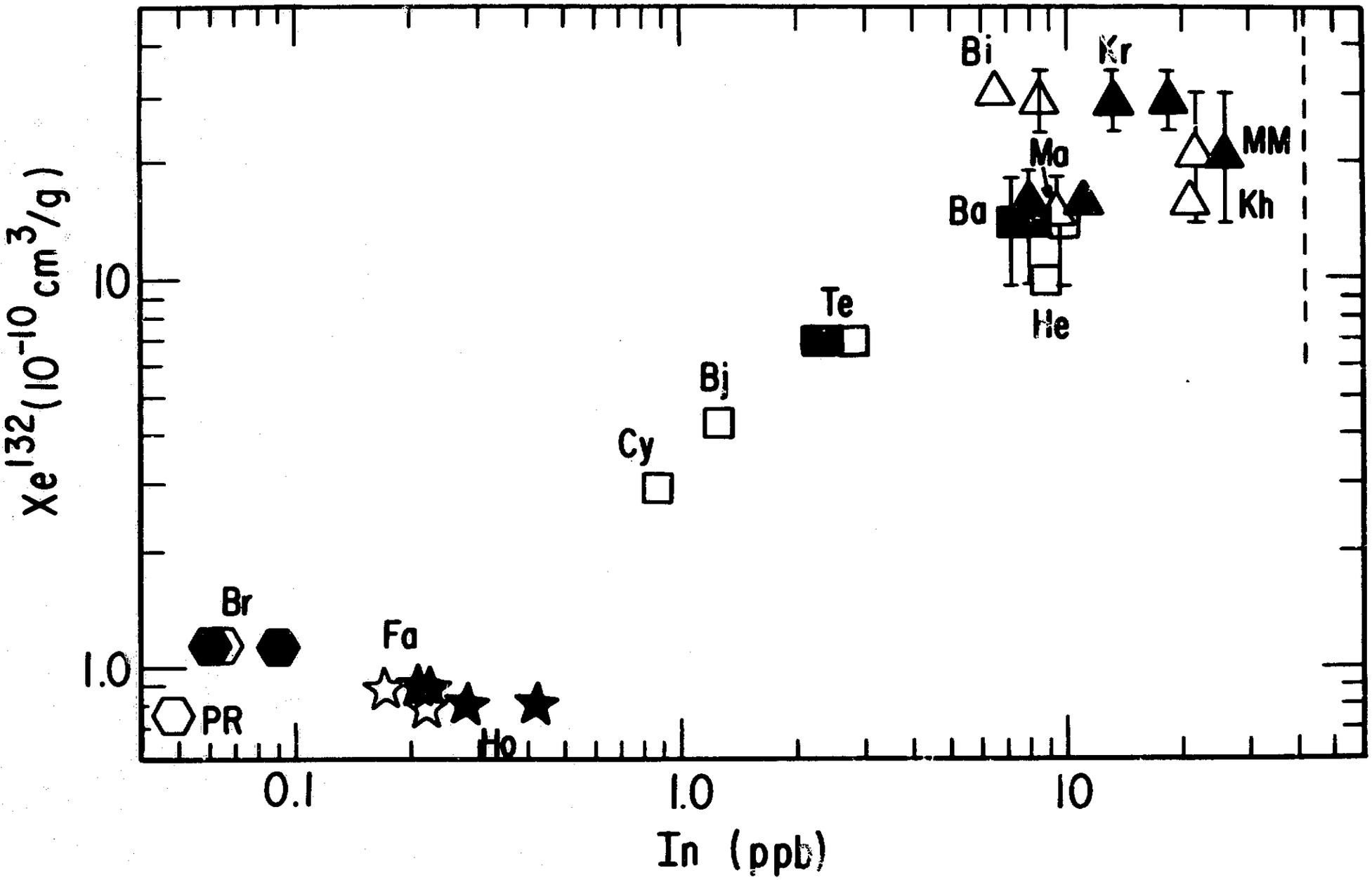

Bi
Kr
Ma
MM
Ba
Kh
He
Te
Bj
Cy
Br
Fa
PR
Ho
10
1.0
0.1
1.0
10
$Xe^{132}(10^{-10}cm^3/g)$
In (ppb)

Fig.5

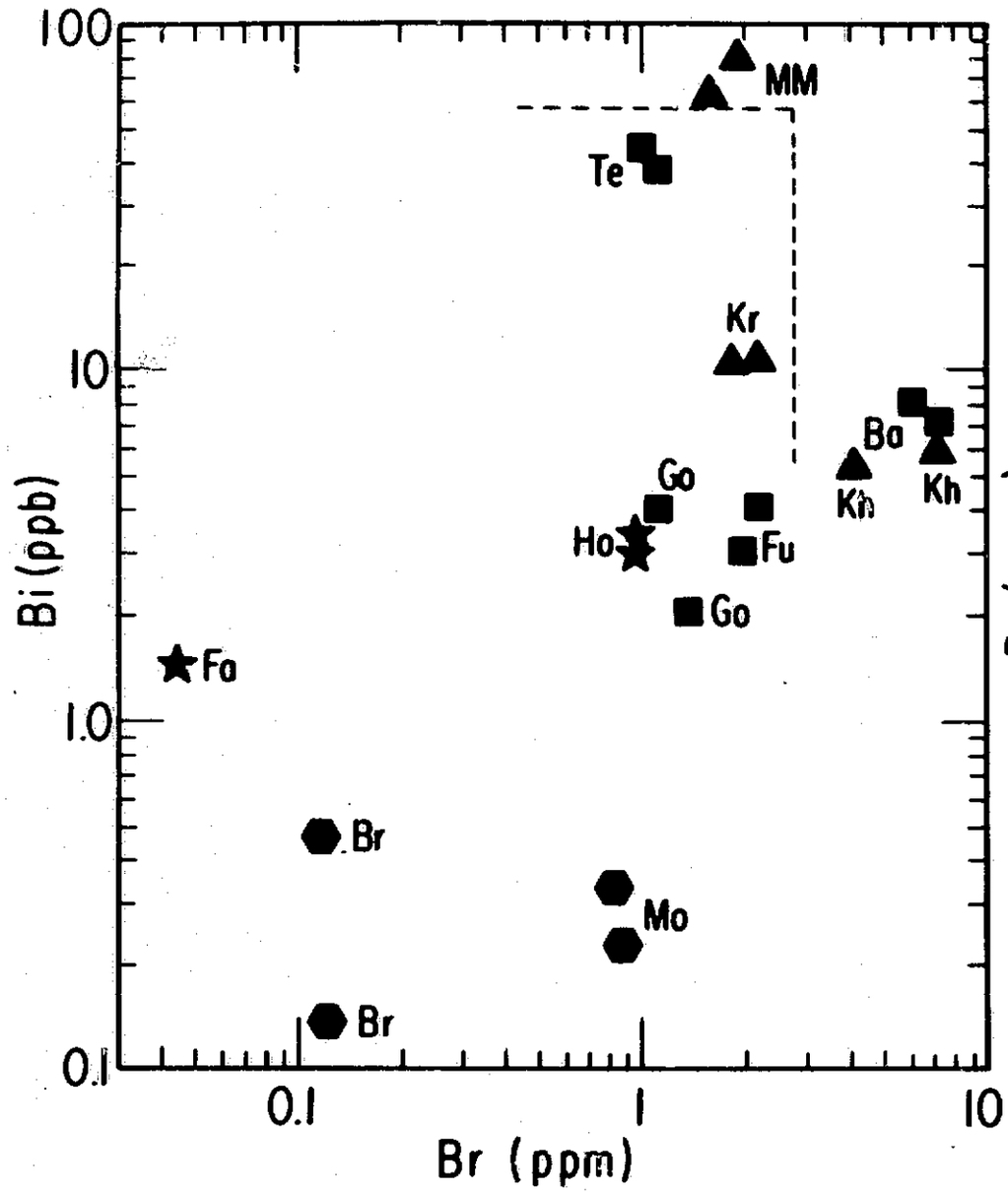

100
10
1.0
0.1
Bi (ppb)
MM
Te
Kr
Ba
Kh
Kh
Go
Ho
Fu
Go
Fa
Br
Mo
Br
0.1
1
10
Br (ppm)

Fig.6(a)

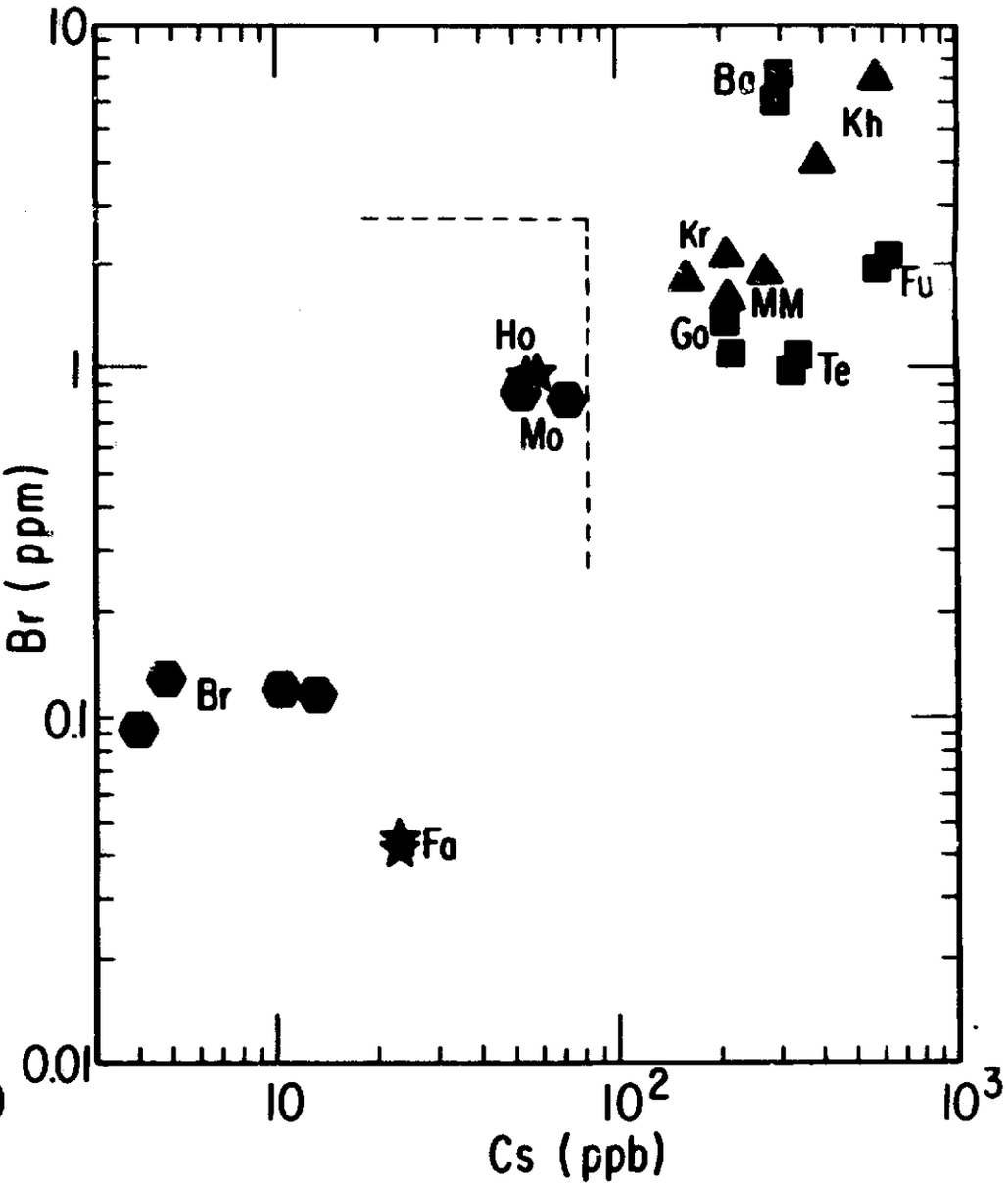

10
1
0.1
0.01
Br (ppm)
Ba
Kh
Kr
Fu
MM
Go
Te
Ho
Mo
Br
Fa
10
10^2
10^3
Cs (ppb)

Fig.6(b)

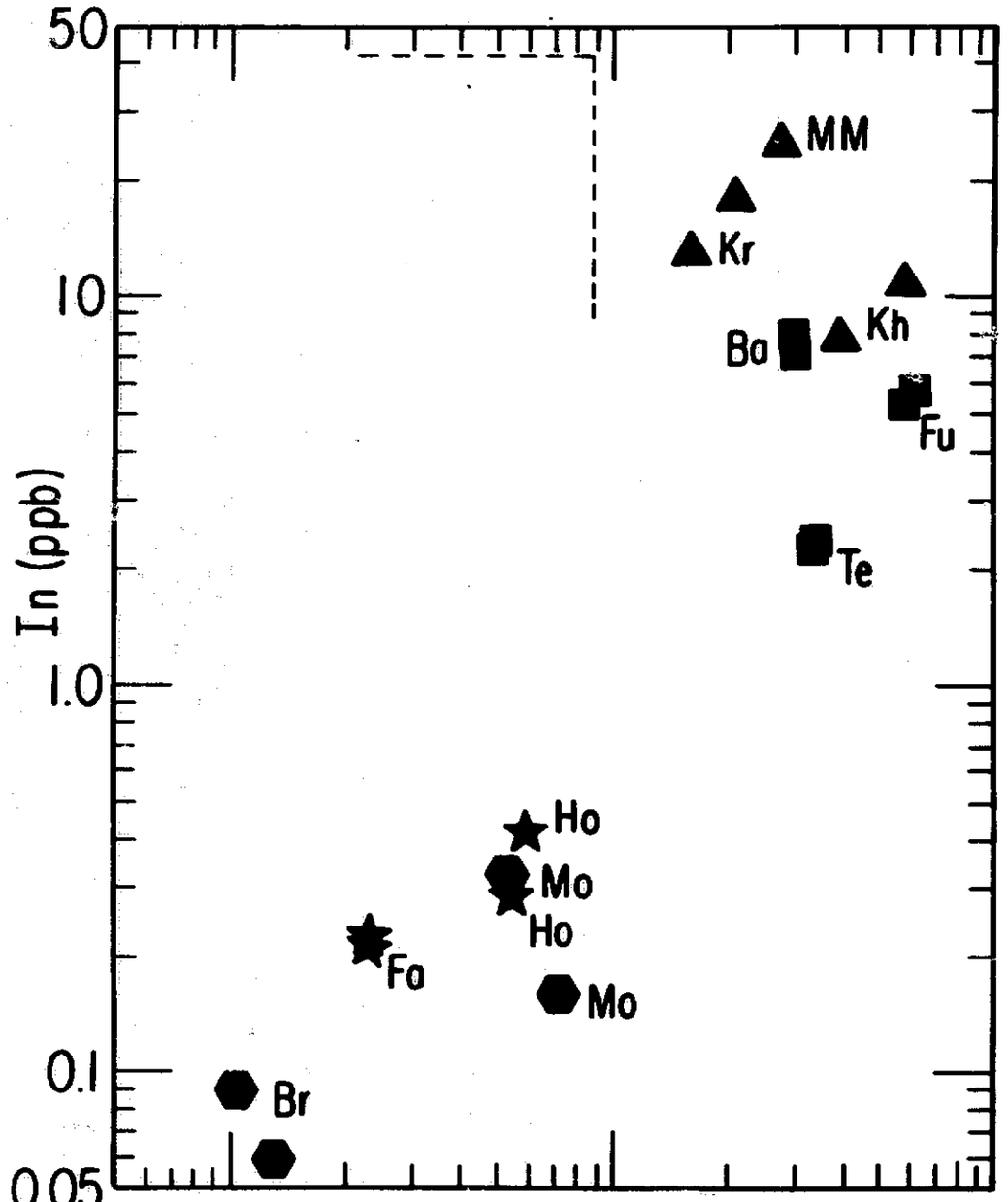

50
10
1.0
0.1
0.05
In (ppb)
MM
Kr
Kh
Ba
Fu
Te
Ho
Mo
Ho
Fa
Mo
Br
10
100
1000
Cs (ppb)

Fig.7(a)

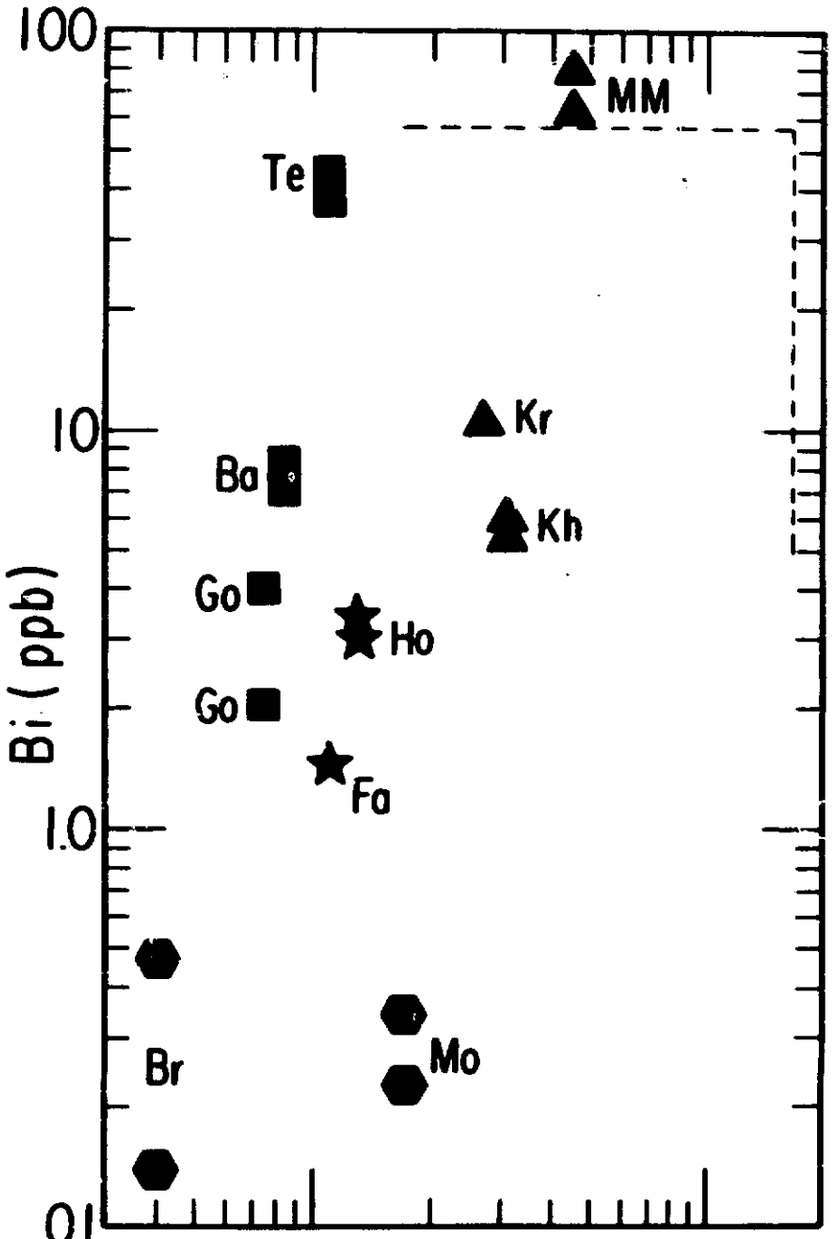

100
10
1.0
0.1
Bi (ppb)
MM
Te
Kr
Ba
Kh
Go
Ho
Fa
Br
Mo
.03
0.1
1.0
2
C (percent)

Fig.7(b)

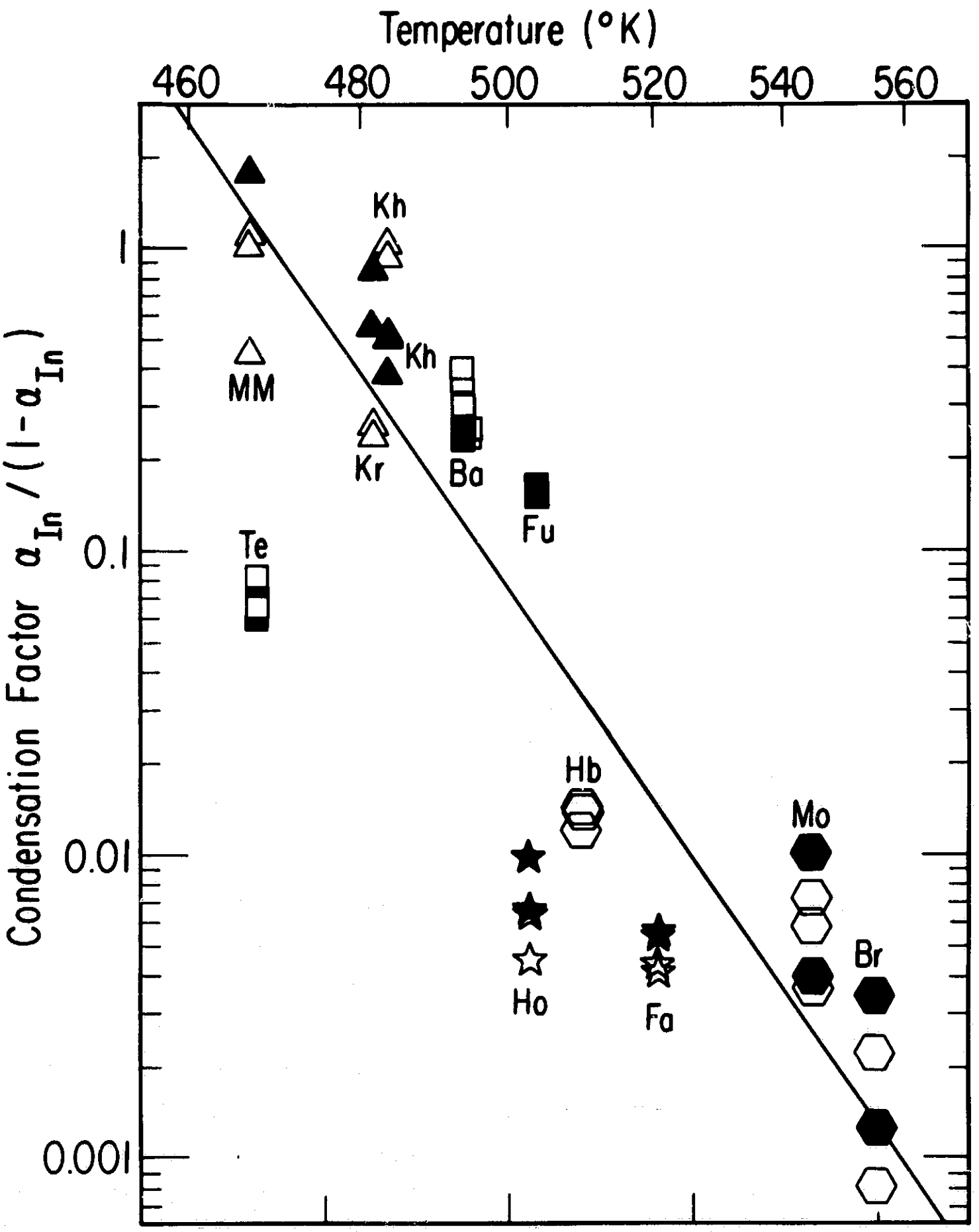

Temperature (°K)
460
480
500
520
540
560
1
0.1
0.01
0.001
Condensation Factor $\alpha_{In}/(1-\alpha_{In})$
Kh
Kh
MM
Kr
Ba
Fu
Te
Hb
Mo
Br
Ho
Fa
2.2
2.1
2.0
1.9
1.8
1000 / T

Fig.8

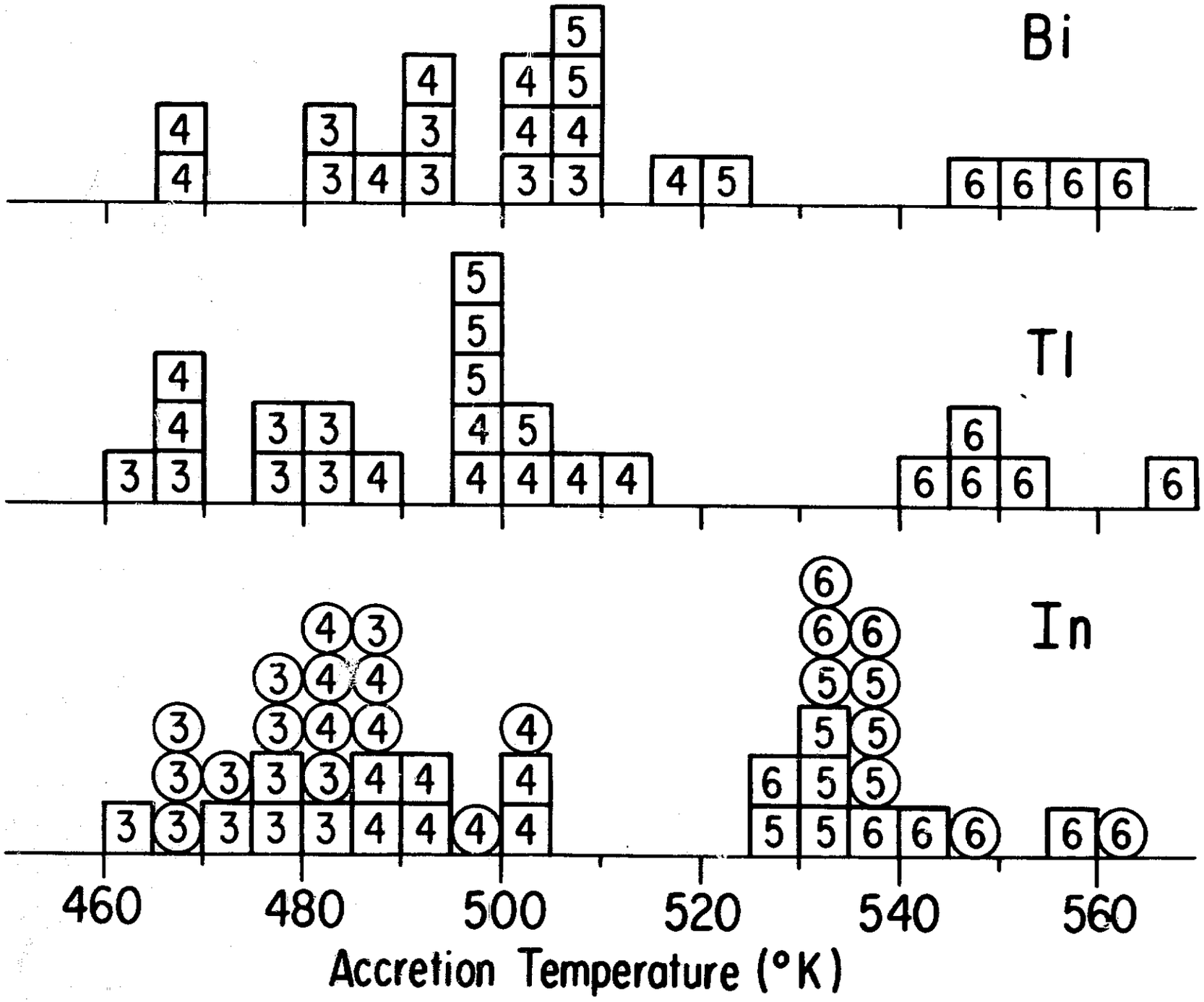


Fig. 9