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Minor and Trace Elements in Meteoritic Minerals

ABSTRACT

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Introduction

During the past decade a very large amount of data has been accumulated on the abundances of minor and trace elements in meteorites, and adequate information is now available on abundance levels of practically all elements for the different classes of meteorite. The distribution of these elements, however, among the individual phases (minerals) of the meteorites is less well known. This can be ascribed in part to the unavailability of large enough samples of many meteorites (especially of the rarer classes) for mineral separation, and in part to the difficulty of making clean separations of the fine-grained minerals in many meteorites, particularly the chondrites. The chondrites are, of course, of special interest, since they represent the least fractionated group of meteorites, and contain many minor and trace elements in amounts comparable to solar abundances.

Some significant information has been obtained by the analysis of magnetic (e.g., metallic) and nonmagnetic (e.g., sulfide plus silicate) fractions of chondritic meteorites (see, for example, Onishi and Sandell (1956), Wardani (1957), Shima (1964), Fouché and Smales (1967). This procedure, however, is useful only for distinguishing the extremely siderophile elements, and provides no information on the distribution of the other elements within the individual minerals. Refined techniques of selective solution of the different phases have been applied to chondrites by

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Vilcsek and Wänke (1965) and Honda and Shima (1967); however, study of their results indicates that this selective solution is seldom completely quantitative. Moss et al. (1967) have developed a selective decomposition procedure, based on heating a meteorite sample in a stream of dry chlorine, which enables measurement of a number of elements in metal, sulfide, and silicate phases.

The introduction of the electron-beam microprobe has made it possible to measure the concentration of many elements in the individual minerals in a polished section of a meteorite, without the necessity of separating the individual minerals by physical or chemical techniques. However, the lower detection limit of the microprobe is about 100 ppm, whereas comparatively few elements are present in the common minerals of meteorites above that level.

The spark source mass spectrometer appears to be an excellent tool with which to attack this problem. It is extremely versatile, being capable of detecting all elements from boron to uranium at concentrations of the order of 0.1 ppm, and considerably less for some elements. Sample requirements are comparatively small: 100 mg is a convenient amount, but good results can be obtained on samples of 10–20 mg. This is very advantageous for chondritic meteorites, in which many elements are concentrated in phases such as phosphates (apatite and/or merrillite) and chromite, present in amounts of less than 1 percent of the bulk meteorite.

Experimental

MINERAL SEPARATION.—As originally planned, the principal objective of this research was to obtain comprehensive data on the distribution of minor and trace

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elements among the minerals of the common chondrites. Preliminary experiments showed that to obtain adequate samples of accessory minerals such as chromite and the phosphates it would be necessary to start with about 100 g of the meteorite. This limited the selection to chondrites available in considerable amounts. Since terrestrial weathering would be a complicating factor, only fresh falls were considered. Ultimately two were selected from the meteorite collection of the Smithsonian Institution, Modoc (NMNH 360), a hypersthene chondrite belonging to the L6 group of Van Schmus and Wood (1967), and St. Severin (NMNH 2608), a hypersthene chondrite (amphoterite subclass) belonging to the LL6 group. General descriptions of these meteorites, with bulk analyses, have been published: Modoc (Mason and Wiik, 1967); St. Severin (Jarosewich and Mason, 1969).

A crust-free piece weighing about 100 g was cut from each of these meteorites. The pieces were broken into smaller fragments which were carefully crushed in a hardened steel mortar until all the material passed through a 10-mesh sieve. Further crushing in a large agate mortar reduced all the material to -100 mesh, except for coarse metal particles, which were removed with a hand magnet. We found the optimum size for mineral separation was the $-200 +325$ mesh fraction, except for the metal, most of which was coarser than 200 mesh, and for which the $-100 +200$ fraction was used.

The $-200 +325$ fraction was sedimented in a large beaker with distilled water to remove dust. Metallic particles were removed with a magnet from the dried fraction, after which this fraction was separated into two portions by centrifuging in pure CH_2I_2 ($D=3.32$). The denser portion consisted largely of olivine and pyroxene, with minor amounts of troilite and chromite; the less dense portion contained the plagioclase and phosphate minerals; both portions still contained a considerable quantity of composite grains.

Pure plagioclase was obtained by extracting a fraction with $D<2.70$ (centrifugation in CH_2I_2-acetone mixture), purification of this fraction with the Frantz Isodynamic Separator, and boiling the final separate in HCl to remove any adhering acid-soluble minerals.

The phosphate minerals were concentrated in the fraction with $D=3.08-3.20$, followed by passage through the isodynamic separator at full power, the phosphate being entirely nonmagnetic. Only small samples of about 20 mg each were recovered.

The portion with $D>3.32$ was further fractionated using Clerici solution of controlled density. Hypersthene concentrated in the 3.32–3.45 fraction, olivine in the 3.50–3.60 fraction; chromite and troilite formed the fraction with $D>4.0$. These fractions were further processed with the isodynamic separator, yielding reasonably pure separates of hypersthene, olivine, troilite, and chromite. The hypersthene and chromite fractions were further treated with hot HCl to remove any adhering olivine and troilite.

The 100–200 mesh magnetic fraction, consisting of metal particles with adhering silicates, was placed in a beaker of acetone and agitated repeatedly in an ultrasonic cleaner. The silicates were separated from the metal by magnetic separation (Modoc), and by centrifugation in Clerici solution (St. Severin).

The purity of these mineral separates was evaluated by microscopic examination. The Modoc fractions were uniformly purer than those from St. Severin, this being conditioned by the somewhat coarser crystallinity of Modoc. Notes on the individual minerals follow.

Modoc olivine: about 99 percent pure; impurity mainly pyroxene; some grains contain minute opaque inclusions, probably chromite; slight limonite staining on many grains.

Modoc hypersthene: about 98 percent pure; impurities are tiny inclusions of plagioclase and admixed grains of diopside.

Modoc feldspar: about 99 percent pure; principal impurity is pyroxene, as tiny inclusions; a few grains show a small amount of opaque dust, probably chromite.

Modoc troilite: about 98 percent pure, impurities are olivine and pyroxene.

Modoc chromite: about 99 percent pure; impurity is mainly pyroxene.

Modoc metal: about 95 percent pure; principal impurities are olivine and pyroxene.

Modoc phosphate: about 99 percent pure; impurities are tiny inclusions of olivine, pyroxene, and plagioclase.

The impurities in the St. Severin separates were essentially the same as the corresponding Modoc separates, but the estimated purity was lower, viz, olivine, 97 percent; hypersthene, 95 percent; feldspar, 96 percent; troilite, 94 percent; chromite, 98 percent; metal, 90 percent; phosphate, 98 percent.

In order to provide comparative material from other classes of meteorites, the following mineral concen-

trates were prepared from the meteorites listed, using techniques similar to those described above:

Olivine: Marjalahti (Fa_{12}), Springwater (Fa_{18}).

Orthopyroxene: Winona (Fs_6), Johnstown (Fs_{27}), Mt. Egerton (Fs_0).

Diopside: Winona.

Pigeonite: Haraiya.

Plagioclase: Winona (An_{10}), Haraiya (An_{90}).

Troilite: Soroti.

Marjalahti and Springwater are pallasites, Johnstown a hypersthene achondrite, Mt. Egerton an enstatite achondrite, Haraiya a eucrite, Soroti a troilite-rich iron. Winona is a peculiar meteorite, having chondritic composition but lacking chondritic structure (Mason and Jarosewich, 1967).

To provide a comparison with terrestrial minerals, olivine, orthopyroxene, and diopside were separated from a peridotite xenolith in a basalt at Camperdown, Victoria, Australia.

ANALYSIS WITH THE SPARK-SOURCE MASS SPECTROMETER. The mass spectrograph used was the M.S.7, manufactured by Associated Electrical Industries Ltd. (now GEC-Elliott Automation). It utilizes double focusing geometry for the ion path and photographic recording of the mass spectrum. The ion source is a pulsed R.F. spark passed between two electrodes, which have been pressed from a 50:50 mixture of sample powder and a graphite/Lu_2O_3 mix. The ion path is maintained at a vacuum of the order of 10^{-8} torr, as higher pressures cause excessive background on the photoplates.

Each mineral separate was ground in an agate mortar until further grinding had no appreciable effect upon the degree of subdivision of the material. Sample homogeneity is one of the major problems when using this analytical technique. The sensitivity of the method is such that a weight of sample of about 0.1 mg is burnt during each analysis and it must be truly homogeneous. Nicholls et al. (1967) fused the sample with a flux to achieve a satisfactory degree of homogeneity. In this study we were dealing with silicate, sulfide, and metal phases and fusion of the meteorite fractions was not feasible owing to the immiscibility of the sulfide and the metal phases with silicate fluxes.

Following Taylor (1965), the finely ground samples were mixed with an equal weight of graphite/Lu_2O_3 mix. The lutetium oxide was present in the graphite at 50 ppm by weight Lu_2O_3 and was the internal standard in the analysis. The geochemical standards G1 and W1 were each equally mixed with the same graphite/Lu_2O_3 mix and sparked under identical conditions. The mass spectrum for each sample was recorded on an Ilford 02 photoplate which was subsequently processed in accordance with the manufacturers' instructions. Fifteen exposures were taken on each plate and exposure was measured in units of charge, as, owing to the fluctuations of the intensity of the ion beam, a fixed time may not give a consistent exposure. The normal procedure when sparking a sample for an exposure range of 1:1000 was for the spark parameters, the repetition rate and length, to be varied such that the recording devices were operated under their optimum conditions. Unfortunately, changing the sparking conditions produces a change in the relative sensitivities of the elements. To avoid this the conditions of the R.F. discharge of the spark were kept constant, the ion beam being attenuated when necessary by the use of an electrical "chopping" device.

The intensity of the lines used for analysis was read on a Jarrall-Ash microphotometer. These intensities were plotted against exposure, and the exposure required to produce a line of a given, fixed intensity was read from this curve. The lutetium 175 line was used as the internal standard and the exposure required to produce the same given intensity was read from a similarly plotted curve. From the standard plates of G1 and W1, graphs were drawn of ppm against $\dfrac{Ex}{E175}$ where Ex is the exposure required to produce a line of given intensity for the isotope X. E175 is the exposure required to produce a line of the same given intensity for ^{175}Lu.

For each sample Ex and E175 were found by reading the photoplate and plotting the intensities against exposure. The concentration of the element of which X is an isotope was then determined from the graphs relating $\dfrac{Ex}{E175}$ to ppm X produced from the G1 and W1 standard plates. Table 1 gives the mass lines used for the estimation of each element reported here.

The detection limit is fixed by the length of exposure of the plate to the ion beam produced from the sample, and also by the proportion of the element in question that is present as the isotope whose line intensity is measured in the analysis. If 20 percent of

TABLE 1.—*Mass lines used for element estimation*

Element	Mass lines	Element	Mass lines
B	11	Sn	117, 118
F	19	Sb	123
Cl	35	Cs	133
Sc	45	Ba	135, 137, 138
Ti	47	La	139
V	51	Ce	140
Cr	53	Pr	141
Mn	55	Nd	143, 146
Co	59	Sm	147, 149
Ni	62	Eu	151, 153
Cu	63, 65	Gd	155, 157, 158
Zn	66, 67	Tb	159
Ga	69, 71	Dy	161, 163
Ge	74	Ho	165
As	75	Er	166, 167
Br	79	Tm	169
Rb	85	Yb	172, 173, 174
Sr	86, 88	Hf	177, 178, 179
Y	89	Pb	208
Zr	90, 91	Th	232
Nb	93	U	238
Mo	95, 98		

TABLE 2.—*Element abundances used as standards*

[ppm by weight]

Element	G1	W1	Element	G1	W1
B	1.5	12	Mo	7.5	0.8
F	700	200	Cs	1.5	0.90
Cl	100	200	Ba	1,050	160
Sc	2.8	34	La	100	11.9
Ti	1,560	6,400	Ce	170	20
V	15	250	Pr	17	3.5
Cr	20	115	Nd	55	12.5
Mn	230	1,300	Sm	9.0	3.4
Co	2.3	45	Eu	1.4	1.1
Ni	1.2	75	Gd	6.9	3.0
Cu	13	115	Tb	0.70	0.60
Zn	45	83	Dy	5.3	4.0
Ga	19	16	Ho	0.50	0.80
Ge	1.1	1.7	Er	1.4	2.08
As	0.7	2.4	Tm	0.20	0.28
Rb	220	22	Yb	1.0	2.1
Sr	260	180	Hf	6.0	2.0
Y	13	26	Pb	49	8
Zr	210	95	Th	50	2.2
Nb	21	10	U	3.2	0.5

an element X is present as one isotope, and this isotope is used in the determination of the concentration of X, for a given exposure the detection limit of X is five times that of a second element Y which has only one isotope. In this work, the detection limit was set at approximately 0.1 ppm atomic for mono-isotopic elements, i.e., the exposure range selected was between $0.1.10^{-9}$ and 100.10^{-9} coulombs.

The accuracy of the figures are dependent upon the values accepted for the geochemical standards used for calibration, and upon the purity of the mineral separates. The latter has been discussed already and the values used for standardization are given in Table 2. One plate was run for each sample and the fifteen graded exposures on each plate gave four or five points on the intensity/exposure graph. The resulting precision is of the order of ± 20 percent.

Elemental Abundances in Separated Minerals

Boron: This element is present in measurable amounts in the feldspar (f) and olivine (o) separates, as follows (in ppm) : Modoc, 2(f) 3(o) ; St. Severin, 3(f) 2(o) ; Winona, 5(f) ; Haraiya, 2(f) ; Marjalahti,

0.5(o) ; Springwater, 3(o). Thus, in meteorites most of this element evidently resides in the feldspar and olivine, as might be predicted from the occurrence of the the boron analogs of these minerals reedmergnerite ($NaBSI_3O_8$) and sinhalite ($MgAlBO_4$).

Fluorine: This element cannot be measured at the low concentrations present in most meteoritic minerals, due to interference from iron ($1/3 Fe^{57}$). In the phosphate concentrates Modoc contained 880 ppm, St. Severin 36 ppm; this remarkable contrast is evidently due to the presence of apatite in the Modoc phosphate and its practical absence in St. Severin.

Chlorine: Chlorine is a major constituent of the Modoc phosphate, the amount being too great to measure with the mass spectrometer. Assuming the composition of chondritic apatite given by Van Schmus and Ribbe (1969), then from the 880 ppm F the amount of chlorine in the Modoc phosphate would be 1.2 percent, and this would contribute 60 ppm to the bulk meteorite. St. Severin phosphate contains only 130 ppm, reflecting the practical absence of apatite. Chlorine was measured in the metal phases (Modoc, 74 ppm, St. Severin, 52 ppm), suggesting the presence of occluded lawrencite (Fe, Ni) Cl_2. It was also found in the troilite (Modoc, 13 ppm; St. Severin, 29 ppm;

Soroti, 39 ppm). It was not detected in olivine; in the remaining separates the use of HCl during separation introduced possible contamination.

Scandium: This element was detected in all the phases (Table 3), but the small amount in the metal can probably be ascribed to silicate contamination. Scandium is essentially lithophile, but shows some chalcophile affinity. In stony meteorites most of this element is contained in the pyroxenes, but the plagioclase and olivine also carry a moderate proportion of the total scandium. Although the abundances in the individual phases of the two chondrites are generally comparable, the variation between specific minerals in the two chondrites is greater than expected, and the sum of the amounts in the individual minerals from St. Severin exceeds the amount measured in a bulk sample of this

meteorite. The figures for the Camperdown minerals are similar to those for the meteoritic minerals.

Titanium: This element was detected in all the phases (Table 4). However, the high concentration in chromite and the possible presence of ilmenite means that even slight contamination by these minerals in the separates will greatly enhance the titanium figures. Contamination is probably responsible for the high and variable figures for plagioclase. Corlett and Ribbe (1967) did not detect Ti at the 100 ppm level in microprobe analyses of terrestrial plagioclases in the An_{0-30} and An_{60-100} composition ranges. However, it can be concluded from Table 4 that the major fraction of titanium in meteorites is contained in pyroxenes; meteoritic pyroxenes contain more titanium than pyroxenes in the terrestrial peridotite from Camper-

TABLE 3.—*Scandium (ppm)*

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene [1]	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			8	13	64				
Modoc	1	8	3	10		6	2	44	[2] 8. 1
St. Severin	0. 5	19	9	36		17	2	30	9
Winona				17	50	16			
Haraiya					40	12			
Marjalahti			3						
Springwater			3						
Johnstown				23					
Mt. Egerton				8					
Soroti		10							

[1] In this and succeeding tables clinopyroxene is diopside in Camperdown and Winona, pigeonite in Haraiya. [2] Schmitt et al. (1964).

TABLE 4.—*Titanium (ppm)*

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			140	350	1, 000				
Modoc	[1] <50	50	100	[2] 1, 000		360	[3] 17, 000	2, 100	700
St. Severin	[1] <50	50	220	[2] 1, 000		650	[3] 19, 000	1, 200	650
Winona				[2] 1, 200	[2] 3, 800	200			[4] 840
Haraiya					[2] 2, 100	340			
Marjalahti			46						
Springwater			40						
Johnstown				1, 200					
Mt. Egerton				120					
Soroti		22							

[1] Jarosewich (unpublished). [2] Microprobe analysis. [3] Bunch et al. (1967). [4] Mason and Jarosewich (1967).

down. The titanium figures for the Modoc and St. Severin troilites are about half those determined by Moss et al. (1967) in troilite from three hypersthene chondrites.

Vanadium: Bunch et al. (1967) showed that much of the vanadium in chondrites is contained in the chromite, and this has been confirmed in our study (Table 5). Contamination by chromite may be responsible for the vanadium recorded in the metal and troilite separates. Clearly the principal carriers of vanadium, apart from chromite, are the pyroxenes, clinopyroxene being richer in this element than orthopyroxene. The contents in the minerals of the terrestrial peridotite from Camperdown are comparable with those for the meteoritic minerals. The low content in the Mt. Egerton pyroxene reflects the chalcophile character of vanadium under the extremely reducing conditions of the enstatite achondrites.

Chromium: Possible contamination with chromite is a serious problem, since 0.1 percent of this mineral will add about 370 ppm Cr to any mineral separate. Many of the data in Table 6 may be too high on this account. The figures for troilite, however, are consistent with those published for iron meteorites and troilite nodules. Smith (1966) reported microprobe analyses for olivine from the Huckitta pallasite (330 ppm), and for olivine from Marjalahti (700 ppm); the latter figure is much higher than our figure. The contrast between the pyroxenes from Modoc and St. Severin may be of real significance and not due to inclusions; Modoc is notably rich in chromite, and therefore its pyroxene may be impoverished in this element.

TABLE 5.—*Vanadium (ppm)*

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			9	55	310				
Modoc	7	4	9	40		12	[1] 4, 000	5	
St. Severin	4	5	14	120		30	[1] 4, 000	12	54
Winona				33	100	5			
Haraiya					74	9			
Marjalahti			10						
Springwater			23						
Johnstown				120					
Mt. Egerton				1					
Soroti		5							

[1] Bunch et al. (1967).

TABLE 6.—*Chromium (ppm)*

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite (percent)	Phosphate	Bulk
Camperdown			370	[1] 2, 800	[1] 9, 000				
Modoc	[2] <10	140	350	[1] 800	[1] 4, 100	370	37	340	[3] 3, 800
St. Severin	[2] <10	270	330	[1] 800	[1] 4, 300	1, 000	38	210	[4] 3, 900
Winona				[1] 1, 000	[1] 4, 800	140			[5] 2, 000
Haraiya					[1] 1, 900	100			
Marjalahti			180						
Springwater			270						
Johnstown				[6] 5, 600					
Mt. Egerton				80					
Soroti		730							

[1] Microprobe analysis.
[2] Jarosewich (unpublished).
[3] Mason and Wiik (1967).

[4] Jarosewich and Mason (1969).
[5] Mason and Jarosewich (1967).
[6] Mason (1963a).

The factors controlling the distribution of chromium between chromite and pyroxene remain to be elucidated. The low content in the Mt. Egerton enstatite reflects the highly reduced state of this meteorite, and is in agreement with the microprobe data of Reid and Cohen (1967).

Manganese: The data in Table 7 were obtained partly by the microprobe, for concentrations above 1000 ppm, and by the mass spectrometer for lower concentrations. The results show that in the ordinary chondrites this element is present in approximately equal amounts in olivine and pyroxene, at the 3000 ppm level. The low figure for the Mt. Egerton

enstatite is characteristic of this mineral from enstatite achondrites and enstatite chondrites. Terrestrial minerals from the Camperdown peridotite have lower manganese contents than those from chondrites.

Cobalt: Over 99 percent of this element in stony meteorites is contained in the nickel-iron, so even a slight contamination of the other phases with metal will add considerably to their cobalt content. This is probably responsible for some of the erratic figures in Table 8, especially the high content in Modoc phosphate, and the somewhat higher content in St. Severin silicates compared to those from Modoc. Cobalt was detected in all phases except the Mt. Egerton enstatite.

TABLE 7.—*Manganese (ppm)*

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown..........			[1] 800	[1] 800	[1] 600				
Modoc...............	[2]<10	150	[1] 3,600	[1] 3,600	[1] 2,000	230	[3] 6,000	580	[4] 2,600
St. Severin...........	[2]<10	290	[1] 3,200	[1] 2,900	[1] 1,900	220	[2] 5,000	520	[5] 2,500
Winona..............				[1] 2,000	[1] 1,400	300			[6] 1.940
Haraiya.............					[1] 7,400	320			
Marjalahti...........			[7] 2,200						
Springwater..........			[7] 2,600						
Johnstown...........				[8] 3,900					
Mt. Egerton..........				[9] 150					
Soroti...............		860							

[1] Microprobe analysis.
[2] Jarosewich (unpublished).
[3] Bunch et al. (1967).
[4] Mason and Wiik (1967).
[5] Jarosewich and Mason (1969).

[6] Mason and Jarosewich (1967).
[7] Mason (1963b).
[8] Mason (1963a).
[9] Reid and Bohen (1967).

TABLE 8.—*Cobalt (ppm)*

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown..........			210	50	28				
Modoc...............	[1] 10,000	58	25	7		1	28	620	[1] 800
St. Severin...........	[2] 12,000	40	45	19		6	33	34	[2] 400
Winona....·..........				3	10	1			[3] 400
Haraiya.............					9	1			
Marjalahti...........			7						
Springwater..........			24						
Johnstown...........				11					
Mt. Egerton..........				n.d.					
Sorot...............		42							

[1] Mason and Wiik (1967).
[2] Jarosewich and Mason (1969).

[3] Mason and Jarosewich (1967).

It is more concentrated in olivine than pyroxene, which agrees with data from terrestrial minerals. Terrestrial minerals, however, normally contain a considerably higher cobalt concentration than meteoritic minerals, and this is illustrated by the figures for the minerals from the Camperdown peridotite.

Nickel: The distribution of this element (Table 9) closely resembles that of cobalt, and the same remarks concerning contamination problems apply. Both troilite and olivine contain appreciable amounts of nickel. The figures for troilite agree with unpublished analyses of the same samples by E. Jarosewich: Modoc, 860 ppm; St. Severin, 760 ppm; Soroti, 510 ppm. Pyroxene contains much less nickel than olivine. The consistently higher figures for nickel in the pyroxene, plagioclase, and chromite separates from St. Severin suggest that these separates are slightly contaminated

with the metal phase. Nickel could not be measured in the phosphate separates because of a concurrent phosphorus line (2 x P^{31}). Terrestrial olivine, orthopyroxene, and clinopyroxene from the Camperdown peridotite show about ten times the nickel concentration of the corresponding meteoritic minerals.

Copper: Hey and Easton (1968) have recently studied the distribution of this element in chondrites by selective decomposition of the different phases. They found 22–65 ppm in kamacite, 1610–2610 ppm in taenite, 1–114 ppm in troilite, 17.8–41.5 ppm in olivine, 6.0–22.3 ppm in pyroxene (plus plagioclase). Our results (Table 10) are in good agreement with these values, although the somewhat erratic figures for troilite and chromite suggest the presence of sporadic grains of native copper. The high figure for St. Severin metal reflects the fact that the nickel-iron in this

TABLE 9.—Nickel (ppm)

Meteorite	Metal (percent)	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			2,000	630	310				
Modoc	[1] 14.9	820	230	46		<20	20	conflict	[1] 13,000
St. Severin	[2] 26.8	910	290	190		74	410	conflict	[2] 10,500
Winona				34	13	27			[3] 12,000
Haraiya					20	<20			
Marjalahti			34						
Springwater			110						
Johnstown				[4] 10					
Mt. Egerton				n.d.					
Soroti		570							

[1] Mason and Wiik (1967).
[2] Jarosewich and Mason (1969).
[3] Mason and Jarosewich (1967).
[4] Mason (1963a).

TABLE 10.—Copper (ppm)

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			7	3	4				
Modoc	620	220	14	7		5	460	52	170
St. Severin	1,900	64	14	10		10	21	20	71
Winona				13	8	7			
Haraiya					35	16			
Marjalahti			3						
Springwater			4						
Johnstown				9					
Mt. Egerton				1					
Soroti		130							

meteorite is entirely taenite, whereas that in Modoc is a mixture of kamacite and taenite. Our figures for the silicates are comparable with theirs, although we find somewhat less in olivine than they did. Pallasite olivine and achondrite orthopyroxene contain less copper than these minerals in chondrites. Terrestrial olivine, orthopyroxene, and clinopyroxene from Camperdown contain less copper than these minerals from chondrites.

Zinc: The data are presented in Table 11. Zinc was not detected in the metal phases. In the analyses of the troilite the Zn^{66} line could not be utilized, because of conflict with 2 x S^{33}, and the Zn^{67} line was absent (an isotope of low abundance); Nishimura and Sandell (1964) report 10±5 ppm Zn in troilite from chondrites, which would be consistent with our data. Zinc is distributed in all the oxidic phases. Chromite may contain a high concentration, as in St. Severin; Bunch et al. (1967) report about 1.1 percent ZnO in one chondritic chromite, that from Pultusk. In the silicates

zinc is about equally concentrated in olivine and pyroxene, with comparatively little in plagioclase. Zinc is very low in pallasite olivine, and in achondrite orthopyroxene.

Gallium: This element was measurable in all the separates except the Mt. Egerton enstatite and the Soroti troilite (Table 12). It is notable for being detectable in all phases, but is concentrated in the metal, plagioclase, and chromite. However, the plagioclase in Winona and especially Haraiya is notably depleted in comparison to the chondrite plagioclases. The lower content in metal from St. Severin may be related to the somewhat higher degree of oxidation of this meteorite compared to Modoc. The figures obtained are consistent with those reported by Moss et al. (1967) and Fouché and Smales (1967) for other chondrites. The concentrations in terrestrial minerals from the Camperdown peridotite are similar to those in the meteoritic minerals.

TABLE 11.—Zinc (ppm)

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			19	6	6				
Modoc	n.d.	n.d.	20	17		3	86	19	
St. Severin	n.d.	n.d.	19	14		3	1,800	22	
Winona				48	20	5			
Haraiya					19	5			
Marjalahti			3						
Springwater			5						
Johnstown				5					
Mt. Egerton				n.d.					
Soroti		n.d.							

TABLE 12.—Gallium (ppm)

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			0.8	1	2				
Modoc	22	6	2	2		17	65	2	
St. Severin	10	5	2	2		23	69	2	5
Winona				1	1	8			
Haraiya					2	3			
Marjalahti			0.6						
Springwater			1						
Johnstown				1					
Mt. Egerton				n.d.					
Soroti		n.d.							

Germanium: This element was found only in the metal phase of the meteorites. Fouché and Smales (1967) found 128 ppm in Modoc metal, and using this figure as a standard we find 280 ppm in St. Severin metal. Since the metal in St. Severin is about half as abundant as in Modoc, the total germanium content of each meteorite is approximately the same, about 9 ppm. Moss et al. (1967) found 122 ppm and 127 ppm in the nickel-iron of the Barwell and Ohuma meteorites, which are hypersthene chondrites similar in composition to Modoc. In the Camperdown minerals germanium was not detected in the olivine, and was present at about 2 ppm in the orthopyroxene and the diopside.

Arsenic: Like germanium, this element has been detected only in the meteoritic nickel-iron; we found 25 ppm in Modoc metal, 43 ppm in St. Severin metal. Both meteorites evidently contain about the same amount of this element, but this is more highly concentrated in the smaller amount of metal in St. Severin. Fouché and Smales (1967), by neutron activation, determined 20.4 ppm in Modoc metal, in good agreement with our figure.

Selenium: This element is concentrated in the troilite, and has not been detected in other phases. The concentration has not been measured because of the lack of suitable standards, but using the average of 8 ppm for chondritic meteorites determined by DuFresne (1960), the amount in the troilites is about 130 ppm.

Bromine: This element was detected in metal, troilite, and phosphate separates, at about the 1 ppm level (except for Modoc phosphate, which contained on the order of 5–10 ppm).

Rubidium: As expected from crystallochemical considerations, essentially all the rubidium is contained in the feldspar; the only separates in which this element was detected were sodic plagioclase from Modoc (28 ppm), St. Severin (5 ppm), and Winona (12 ppm). It was not detected in the calcic plagioclase from Haraiya. It is remarkable that the level of Rb in St. Severin feldspar is only about one-fifth that of Modoc; however, the Rb content of the bulk meteorite (0.8 ppm) is also much below the chondrite average. Smales et al. (1964) have found that the rubidium content of different chondrites is notably erratic.

Strontium: The results reported in Table 13 show that the plagioclase contains most of the strontium in these meteorites, although this element is also enriched in the phosphate separates. Pyroxene contains more strontium than olivine. This element was not detected in the metal or troilite separates.

Yttrium: Table 14 shows that yttrium, like the lanthanides, is largely contained in the phosphate minerals. Diopside from Winona and pigeonite from Haraiya show a concentration of this element. Otherwise, detectable amounts were found in orthopyroxene, plagioclase, and in the troilite. The amounts in terrestrial orthopyroxene and diopside are somewhat lower than in the meteoritic minerals.

Zirconium: From crystal chemistry considerations, this element is unlikely to substitute readily for any of the major cations. The mineral zircon ($ZrSiO_4$) has been observed in a few mesosiderites and one chondrite, and sporadic grains of this mineral would readily account for the amount of zirconium (of the order of 10 ppm) in stony meteorites. The general consistency of the data for individual minerals in Table 15, how-

TABLE 13.—Strontium (ppm)

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			1	4	280				
Modoc	n.d.	n.d.	0.4	4		75	8	87	11
St. Severin	n.d.	n.d.	0.9	5		73	6	62	10
Winona				5	14	73			
Haraiya					6	130			
Marjalahti			0.5						
Springwater			0.4						
Johnstown				2					
Mt. Egerton				2					
Soroti		n.d.							

TABLE 14.—*Yttrium* *(ppm)*

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			n.d.	0. 5	9				
Modoc	n.d.	1	n.d.	0. 8		<1	n.d.	160	[1] 2. 07
St. Severin	n.d.	1	n.d.	2		<1	n.d.	210	2
Winona				1	23	<1			
Haraiya					15	1			
Marjalahti			n.d.						
Springwater			n.d.						
Johnstown				1					
Mt. Egerton				n.d.					
Soroti		0. 5							

[1] Schmitt et al. (1964).

TABLE 15.—*Zirconium* *(ppm)*

Meteorite	Metal	Troilite	Olivine	Ortho-pyroxene	Clino-pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			1	3	60				
Modoc	n.d.	3	n.d.	6		7	5	650	8
St. Severin	n.d.	3	n.d.	22		8	2	18	6
Winona				6	90	4			
Haraiya					29	2			
Marjalahti			n.d.						
Springwater			n.d.						
Johnstown				2					
Mt. Egerton				n.d.					
Soroti		3							

ever, suggests that sporadic zircon is not responsible for the distribution pattern (except probably for the very high figure in Modoc phosphate, and possibly for the Winona diopside and St. Severin orthopyroxene). Most of the zirconium appears to be present in the calcium-rich clinopyroxene, a trend followed also by the terrestrial minerals in the Camperdown peridotite. The presence of 3 ppm in troilite indicates minor chalcophile affinity for zirconium, as has been suggested by Setser and Ehmann (1964); they recorded 6.3 ppm in troilite from the Canyon Diablo iron.

Niobium: The concentration of niobium in ordinary chondrites is not well known, but is probably of the order of 1 ppm. We have measured 1–2 ppm in meteoritic chromite, phosphate, and clinopyroxene, and detected this element (<1 ppm) in troilite, olivine, orthopyroxene, and plagioclase. The Camperdown

minerals contain niobium at approximately the same concentration as the corresponding meteoritic minerals.

Molybdenum: This element was observed in metal and troilite separates, with somewhat greater concentration in the metal; it was not observed in other phases. Results obtained (in ppm) are: Modoc metal, 7; Modoc troilite, 3; St. Severin metal, 10; St. Severin troilite, 3; Soroti troilite, 3. These figures are consistent with the data of Kuroda and Sandell (1954), although their results are generally somewhat larger.

Ruthenium, rhodium, palladium: These three geochemically coherent elements were prominently present in spectra of the metal phases only. Previous suggestions that ruthenium and possibly palladium show some chalcophile affinity in meteorites were not borne out; neither element was detected in troilite. The amounts of these elements in the metal phases

could not be evaluated because of the lack of suitable standards. Nichiporuk and Brown (1965) reported the following average amounts in chondrite metal: Ru, 6 ppm; Rh, 1 ppm; Pd, 4 ppm. Fouché and Smales (1967) report 6.87 ppm in Modoc metal.

Silver: This element was just visible in spectra of the troilites, but absent from those of other phases.

Cadmium: This element was not detected.

Indium: This element was not detected.

Tin: Tin was detected only in the metal phase; 2 ppm in Modoc metal, 8 ppm in St. Severin metal. This is in agreement with earlier work by Onishi and Sandell (1957) and Shima (1964), who found that this element is essentially confined to the magnetic (i.e., metallic) fraction of chondritic meteorites.

Antimony: This element was found at about 1 ppm in the metal phase only. Fouché and Smales (1967) record 0.72 ppm in metal from Modoc.

Tellurium: Lines of tellurium are visible in spectra of the metal and troilite, and are absent in spectra of the other phases. The concentration is approximately the same in the metal and in the troilite. This is contrary to the results of Goles and Anders (1962) on iron meteorites; they record 5 ppm in Canyon Diablo troilite, 0.09 ppm in the metal; and 1.7 ppm in Toluca troilite, 0.05 ppm in the metal. The equilibrium conditions in iron meteorites, however, may be considerably different for this element from those in the chondrites.

Iodine: Since methylene iodide was used in the mineral separations (except for the Soroti troilite) the iodine line observed in these separates can be ascribed to contamination. Iodine was not detected in

the Soroti troilite. Goles and Anders (1962) record that this element is chalcophile in iron meteorites; they found 0.06 ppm in Canyon Diablo troilite, 0.03 ppm in the metal, and 1 ppm in Toluca troilite, 0.25 ppm in the metal.

Cesium: This element was detected in plagioclase from Modoc (0.4 ppm), St. Severin (0.3 ppm), and Winona (0.4 ppm).

Barium: The results (Table 16) show, as might be predicted, that most of the barium in a stony meteorite is contained in the feldspar. The phosphate contains a moderate amount, as does the Winona diopside. The other minerals contain about 1–3 ppm barium, except the metal, in which this element was not detected.

Lanthanides: The data for the lanthanides (here considered to comprise the elements La-Lu) are given in Table 17 and illustrated in Figure 1. In the chondrites (Modoc and St. Severin) these elements are highly enriched in the phosphate minerals, and are at concentrations of less than about 0.2 ppm in the other phases (except europium, which is present in the feldspar at about 1 ppm). Comparison of the data for the phosphate minerals with those for a chondrite composite (Haskin et al., 1968) shows that the phosphates account for almost 100 percent of the lighter lanthanides (La-Sm), and 50 percent or more of the heavier lanthanides (Gd-Yb). The amounts unaccounted for in the phosphates are probably contained in the pyroxenes, which consistently show an enrichment in the heavy relative to the light lanthanides.

The Haraiya separates, calcic plagioclase and

TABLE 16.—Barium (ppm)

Meteorite	Metal	Troilite	Olivine	Ortho- pyroxene	Clino- pyroxene	Plagioclase	Chromite	Phosphate	Bulk
Camperdown			1	3	2				
Modoc	n.d.	1	<0.5	2		42	2	11	[1] 3.8
St. Severin	n.d.	1	0.8	2		40	0.8	16	6
Winona				3	8	51			
Haraiya					1	72			
Marjalahti			0.9						
Springwater			3						
Johnstown				0.6					
Mt. Egerton				1					
Soroti		1							

[1] Reed et al. (1960).

TABLE 17.—*Lanthanide elements (ppm)*

Element	Modoc Phosphate	St. Severin Phosphate	Winona			Haraiya		Angra dos Reis
			Feldspar	Enstatite	Diopside	Feldspar	Pyroxene	
La.....	51	64	0. 94	0. 13	3. 0	1. 4	0. 25	8. 3
Ce.....	120	130	0. 84	0. 33	11	2. 1	0. 86	19
Pr......	17	25	0. 093	0. 080	2. 1	0. 22	0. 19	3. 7
Nd.....	65	100	0. 30	0. 41	11	0. 82	0. 95	17
Sm.....	27	34	<0. 15	<0. 2	5. 1	0. 30	0. 51	5. 5
Eu.....	2. 5	2. 5	0. 51	<0. 04	0. 085	0. 66	<0. 02	1. 6
Gd.....	30	50	<0. 13	0. 34	7. 2	0. 60	0. 80	7. 6
Tb.....	4. 4	6. 8	<0. 03	0. 040	0. 83	0. 06	0. 12	1. 1
Dy.....	34	49	<0. 14	0. 33	6. 5	0. 43	1. 1	8. 7
Ho.....	8. 6	12	<0. 04	0. 090	1. 6	0. 075	0. 31	2. 0
Er......	19	26	<0. 10	0. 24	3. 1	0. 20	0. 85	4. 6
Tm....	2. 4	3. 2	<0. 02	0. 04	0. 32	<0. 04	0. 13	0. 56
Yb.....	18	25	<0. 10	0. 33	2. 4	0. 22	1. 0	4. 2

pigeonite, show the preference of the lighter lanthanides and europium for the plagioclase and that of the heavier lanthanides for the clinopyroxene, a phenomenon already estabilshed for the Juvinas eucrite (Philpotts et al., 1967) and for terrestrial plagioclase/pyroxene pairs. The Winona separates (plagioclase, enstatite, and diopside) show the same trend but with a larger separation of their lanthanide patterns. The considerable enrichment of all the lanthanides in diopside relative to enstatite (and relative to Haraiya pigeonite) is evidently related to the crystal structure of diopside, with a lattice position which readily accommodates calcium and ions of similar size, such as the lanthanides. Although the lanthanides are highly concentrated in diopside relative to enstatite, these two minerals account for comparable amounts of each element in the meteorite as a whole, since the enstatite is much more abundant (40 percent) than the diopside (2.5 percent).

The Angra dos Reis meteorite, which consists almost entirely of augite, has a lanthanide pattern that is highly enriched and very little fractionated when compared with that of chondritic meteorites. Schnetzler and Philpotts (1969) report comparable results for some of these elements. Angra dos Reis show only slight improverishment in europium, unlike the other pyroxene separates. This suggests either that the conditions of crystallization were such that no Eu^2 could form and that the element behaved as any other triply charged lanthanide, or that there was no competing phase, such

as plagioclase, that would capture any Eu^2 were it present.

Hafnium: As might be predicted, hafnium follows the pattern of zirconium, being detected only in those samples relatively rich in this element—St. Severin orthopyroxene and phosphate, Modoc phosphate, and clinopyroxene from Winona, Haraiya, and Camperdown peridotite. The concentration in Modoc phosphate is 18 ppm, in Winona diopside 4 ppm; in the other samples the concentration was too low for accurate measurement.

Tantalum: In view of its low abundance, it is unlikely that this element could be detected in any of the meteorite mineral separates. In addition, the tantalum mountings of the spark source are a possible contaminant.

Tungsten, rhenium: These elements were detectable in the metal phases, but were not observed in any of the other separates. Fouché and Smales (1967) report 0.52 ppm Re in Modoc metal.

Osmium, iridium, platinum, gold: The lines of these elements are prominent in the spectra of the nickel-iron, but are absent from those of the other separates. Suitable standards for evaluating the amounts were not available, but the results are consistent with the figures given for chondrite metal by other investigators: Ir, 2.8 ppm (Nichiporuk and Brown, 1965); Pt, 8.5 ppm (Nichiporuk and Brown, 1965); Au, 1.2 ppm (Baedecker and Ehmann, 1965). Fouché and Smales (1967) report 2.11 ppm Au in Modoc metal.
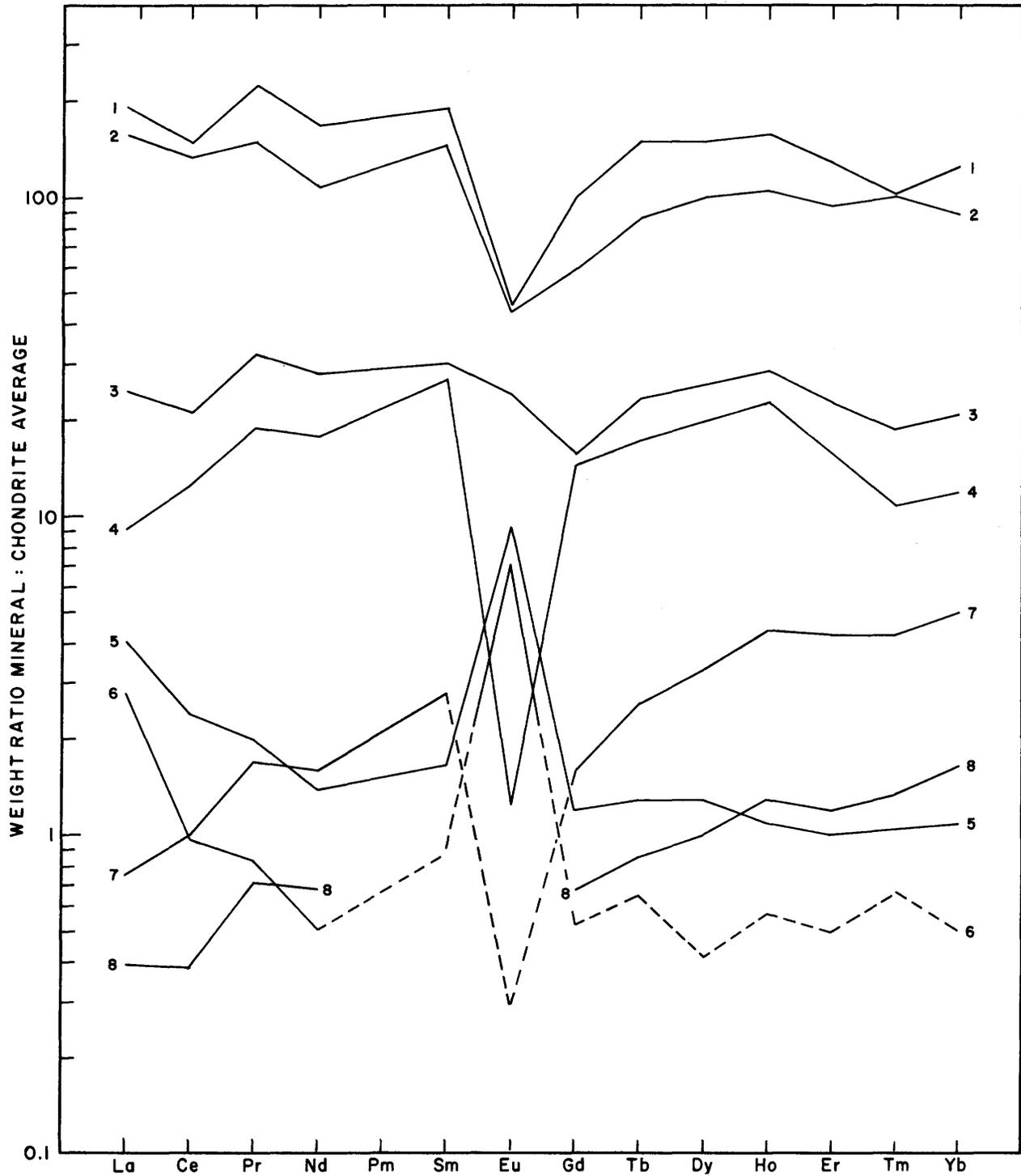
FIGURE 1.—Chondrite-normalized lanthanide concentrations in meteorite minerals (broken lines indicate maximum values): 1, St. Severin phosphate; 2, Modoc phosphate; 3, Angra dos Reis; 4, Winona diopside; 5, Haraiya plagioclase; 6, Winona plagioclase; 7, Haraiya pigeonite; 8, Winona enstatite. (Note added in proof: The points for Gd are systematically low, the values used being one-half those given for this element in Table 17.)

Mercury: This element was not detected.

Thallium: This element was not detected, except in those separates for which Clerici solution had been used as a separating medium, thereby introducing thallium as a contaminant.

Lead: This element was detected in both metal and troilite separates from Modoc, with the troilite having the greater concentration. The metal and troilite separates from St. Severin also showed lead, but since these had been treated with Clerici solution, this element may have been thereby introduced. Lead is present in Soroti troilite at a concentration of 4 ppm. Lead was also detected in Haraiya and Winona feldspar.

Bismuth: This element was not detected in any separates.

Thorium: Only the phosphate separates contain detectable amounts of this element, Modoc phosphate having 7 ppm, St. Severin phosphate 6 ppm.

Uranium: This element was barely detectable in St. Severin phosphate, and was measured at 4 ppm in the Modoc phosphate. The latter figure seems somewhat high, in view of the known Th:U ratio.

Summary

Although the number of meteorites examined in this research is small, they are representative of the major classes of stony meteorites, except the enstatite and carbonaceous chondrites. The following generalizations regarding the distribution of minor and trace elements in specific minerals, therefore, may be tentatively offered.

Nickel-iron: The metal phase in an ordinary chondrite contains over 99 percent of the nickel and cobalt in the meteorite, and probably at least 90 percent of the following elements: Ge, As, Ru, Rh, Pd, Sn, Sb, W, Re, Os, Ir, Pt, Au. Copper and gallium show both siderophile and lithophile affinity, and molybdenum and tellurium show both siderophile and chalcophile affinity.

Troilite: It is noteworthy that the only measured element which is entirely chalcophile, appearing in troilite but in no other phase, is selenium. Silver, cadmium, indium, thallium, and lead are probably concentrated in troilite, although their abundance level was too low for measurement. Elements present in troilite at the 100–1000 ppm level are Ni, Cr, Mn, and Cu; at the 10–100 ppm level Cl, Sc, Ti, and Co; at the 1–10 ppm level V, Zn, Ga, Y, Zr, Mo, and Ba.

Olivine: This mineral does not readily accept minor and trace elements into its structure. Of the elements studied only Mn is present in meteoritic olivine in notable amounts (2000–3000 ppm). Meteoritic olivine contains more Ni and Co than other meteoritic silicates, but the concentration of these elements is about an order of magnitude lower than in terrestrial olivine such as that in the Camperdown peridotite. Chondritic olivine contains about 100–200 ppm Ti and about 300 ppm Cr; it contains about 20 ppm Zn, which may represent about half the zinc in the meteorite.

Pyroxenes: Apart from Ni and Co, which are preferentially accepted in olivine, many of the minor and trace elements with lithophile affinity tend to concentrate in the pyroxenes. This is particularly true of the transition elements scandium through manganese. In general, the concentration in clinopyroxene is greater than in orthopyroxene. In chondrites the major part of the lanthanides (and yttrium) are contained in the phosphate phases, but detectable amounts are present in the pyroxenes.

Plagioclase: As might be predicted, the sodic plagioclase of chondrites contains most of the Rb, Cs, Sr, and Ba in these meteorites. It also contains appreciable amounts of the Sc and Ga.

Chromite: Apart from chromium, this mineral also concentrates vanadium, and to a lesser extent titanium, manganese, and gallium.

Phosphate: The phosphate minerals in chondritic meteorites account for most of the lanthanides, yttrium, uranium, and thorium in these meteorites. If apatite is present, it will contain essential amounts of fluorine and chlorine. Other elements enriched in the phosphate minerals include Sc, Sr, Zr, Ba, and Hf.

Acknowledgments

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