HETEROGENEITIES IN THE SOLAR NEBULA

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204

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Abstract

Oxygen isotopic compositions of the high-temperature phases in carbonaceous chondrites define a mixing line with an ^{16}O-rich component and show little superimposed chemical isotope fractionation. Within a single inclusion in Allende, variations of $\delta^{18}O$ and $\delta^{17}O$ of 39‰ are found. The ordinary chondrites are slightly displaced from the terrestrial fractionation trend, implying that at least 0.2% of the oxygen in terrestrial rocks was derived from the ^{16}O-rich component.

Introduction

It was recently shown that the anhydrous phases of the carbonaceous chondrites of classes C2 and C3 display a remarkable pattern of oxygen isotope abundances: an almost constant ratio of $^{18}O/^{17}O$ and highly variable ^{16}O (Clayton et al., 1973). The variations of $^{18}O/^{16}O$ and $^{17}O/^{16}O$ are much greater than can be accounted for by chemical fractionation processes, and do not conform to a 2:1 ratio as would be expected for any mass-dependent process. It was concluded that the variations in the C2 and C3 meteorites are ultimately nuclear in origin, and represent the mixing of two kinds of oxygen with different nucleosynthetic histories, and hence different isotopic compositions. One component of the mixture is here referred to as "ordinary oxygen" and has isotopic abundances near the values of the mean earth, moon and ordinary chondrites; the other component is highly depleted in the heavy isotopes, and may be nearly pure ^{16}O. Several lines of evidence suggest that the ^{16}O-rich component existed as solid grains of interstellar dust prior to condensation of solids containing "ordinary oxygen" from the solar nebular gas.

The purpose of this paper is to present new data on meteorites, obtained from refined analytical techinques which yield an order of magnitude better precision in $^{17}O/^{16}O$ determinations than those previously used. It is now possible to examine the small isotopic differences among the earth, moon and ordinary chondrites, and to distinguish between the effects of admixture of ^{16}O-rich components into these bodies and the effects of chemical isotope effects during condensation and accretion (Onuma et al., 1972a). It will be shown that even a surficial sample of a differentiated planet (e.g. Mars) can be used to determine the content of ^{16}O-rich component in the planet - a parameter that may be important in understanding planetary accretion.

Analytical Procedures

Sampling: In the equilibrated ordinary chondrites and in the achondrites, isotopic equilibration has occurred in metamorphic and igneous processes, respectively, so that the internal isotopic variations which remain are just the chemical isotopic fractionations between minerals which reflect the maximum temperature of heating (Onuma et. al., 1972b). Sampling of such meteorites presents no special problems.

In the unequilibrated chondrites, and especially in the carbonaceous chondrites, large isotopic variations are present at least down to a sub-millimeter size range, and information is lost in analyzing larger samples. Hence, our emphasis has been on individual chondrules, crystals, or small clusters of crystals, particularly as exemplified by the refractory aggregates in the C3 meteorite, Allende.

Samples, typically 3-10 mg in size, were treated by the bromine pentafluoride technique (Clayton and Mayeda, 1963), and the oxygen thus liberated was analyzed, as O_2, using a 15 cm, 60^0 sector, double-collecting mass spectrometer. Both $^{18}O/^{16}O$ and $^{17}O/^{16}O$ ratios were determined. The results are reported in the δ-notation, relative to SMOW standard (Craig, 1961). Since SMOW is a fictitious standard, defined to have an $^{18}O/^{16}O$ ratio (R^{18}) fixed relative to a real substance, NBS-1 water, by the equation:

$$R^{18}_{SMOW} = 1.008\ R^{18}_{NBS-1} \quad \text{(Craig, 1961)},$$

we have adopted a similar definition for R^{17}_{SMOW}, as follows:

$$R^{17}_{SMOW} = (1.008)^{1/2}\ R^{17}_{NBS-1}$$

Analytical uncertainties are estimated as $\pm$ 0.1‰ both for δ^{18} and δ^{17}.

Results

Oxygen isotopic compositions of some terrestrial and meteoritic samples are given in Table 1. Data from terrestrial samples and the Allende meteorite are shown graphically in Figure 1. It is seen that all of the terrestrial samples lie along a line through the origin with a slope of +1/2. This is to be expected for any samples derived from an initially homogeneous reservoir, in which all subsequent isotopic fractionations are mass-dependent, and conform approximately to the relationship:

$$\alpha^{17}_{AB} = (\alpha^{18}_{AB})^{1/2} \tag{1}$$

where α_{AB} is defined as a fractionation factor between two phases, A and B:

$$\alpha^{17}_{AB} = (R^{17}_{A}/R^{17}_{B}) \text{ and} \tag{2}$$

$$R^{17}_{A} = (O^{17}/O^{16})_{A}. \tag{3}$$

Equation (1) does not hold rigorously for isotopic fractionation processes, but is a good approximation. For example, if isotopic fractionation is brought about by diffusion, with diffusion rates inversely proportional to the square-root of the masses of the diffusing molecules, equation (1) is in error by only about 1% if the diffusing species are oxygen atoms, and is an increasingly better approximation for oxygen-containing molecules of higher molecular weight. For isotopic fractionation in a chemical equilibrium process, equation (1) implies a linear relationship between free energy and mass difference, which is a good approximation for small relative mass differences.

In the paper of Clayton _et al._, (1973), data points for the C2 and C3 meteorites were scattered about the mixing line with deviations from the line on the order of 1‰. It was not clear how much of the scatter was due to analytical uncertainty in the $^{17}O/^{16}O$ measurements, and how much was attributable to chemical isotope fractionations superimposed on the mixing process. It is evident from the new data plotted in Figure 1 that the scatter was principally analytical, and that chemical shifts from the mixing line are small, and probably <0.5‰ for all of the Allende samples plotted. Onuma et al., (1972a) have discussed the magnitudes of the oxygen isotope effects expected for solids condensing from the solar nebula, in which the principal reservoir of oxygen should be carbon monoxide at the high temperatures involved. The small variability of the chemical isotope shifts implies a narrow range of condensation temperatures, and is compatible with the range from 1625° to 1450°K given by Grossman (1972) for the interval between condensation of melilite and condensation of forsterite.

Fig. 2 shows data for other C2 and C3 meteorites, with the lines from Fig. 1 given for reference. It is seen that the points fall on the line defined by the Allende samples. The smaller spread in the data may reflect the sampling technique, which tended to homogenize the samples. It should be noted, however, that the samples from Ornans are all iron-bearing olivines, and it may be surprising that the introduction of lower-temperature ferrous iron did not result in a chemical displacement of the data points from the line.

It is of interest to determine whether the various minerals within a single chondrule or inclusion have crystallized together. A large (1 cm diameter), coarse-grained, round inclusion from Allende was crushed, and mineral concentrates were prepared by heavy-liquid separation. Four fractions, labeled Al-15-1 to Al-15-5 in Table 1, showed a range of 39‰ in δ^{18} and δ^{17}, falling along the mixing line rather than a fractionation line, as would be expected if the minerals had crystallized from a melt.

The densest fraction, composed primarily of titanian clinopyroxene and included spinel, is the most ^{16}O-rich of any sample yet analyzed: the ^{16}O-rich component makes up at least 5% of its oxygen. Even at this high concentration, the physical nature of the ^{16}O-rich component is not revealed by microscopic examination.

Clayton et al., (1973) showed that samples from the moon and the ordinary chondrites fall on or near the terrestrial fractionation line, implying that these bodies have the same fraction of the ^{16}O-rich component, and might, in fact, be composed entirely of "ordinary oxygen". With the increased precision now available, it can be seen in Fig. 3 that analyses of two ordinary chondrites, Pantar (H5) and Modoc (L6) define a fractionation trend parallel to, but displaced from, the terrestrial line. The range of data points from Pantar results from a set of partial fluorination reactions, such as were first carried out on lunar soils (Epstein and Taylor, 1971). The variation of about 4‰ in $\delta^{18}O$ is greater than would be expected for fractionation among the major minerals in a meteorite of petrographic grade 5 (Onuma et al., 1972b), and probably results from prior exposure of the constituents of this gas-rich meteorite to bombardment while in the regolith of the parent body.

If the apparent displacement of the ordinary chondrite data from the terrestrial fractionation line is supported by subsequent analyses, the implications for accretion of the earth are significant. Since the ordinary chondrites are displaced in the ^{16}O-depleted direction, the amount of ^{16}O-rich component in the earth cannot be zero. The magnitude of the displacement sets a lower limit on the content of this component at 0.2% of the oxygen of all terrestrial rocks.

The three-isotope plot for oxygen should be useful in future studies of samples returned from other planets. If the two-component model of solar system oxygen is valid, it is possible to determine the relative proportions of the components in a planet on the basis of a single analysis of a sample of the planet, even if the planet has undergone extensive differentiation, so that the surface sample is not chemically representative of the body as a whole. It would simply be necessary to plot the data point, and construct a fractionation line with slope of 1/2 to find its intersection with the mixing line. The intersection then determines the content of ^{16}O-rich component relative to the earth and the various meteorite classes.

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TABLE 1

Oxygen Isotopic Compositions of Terrestrial and Meteoritic Samples

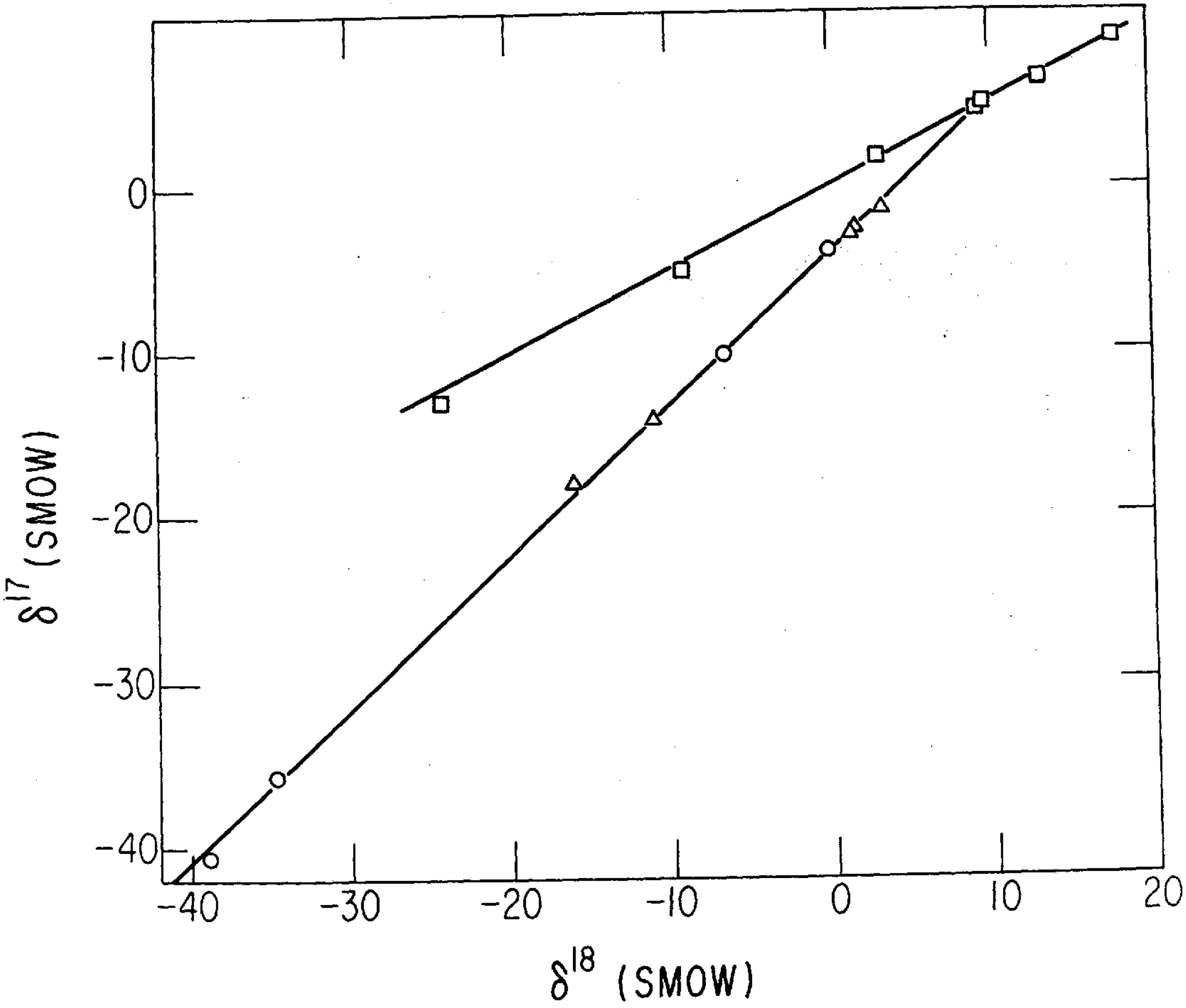
	Sample No.	Description	$\delta^{18}O$	$\delta^{17}O$
Terrestrial	NBS-1a	Yellowstone snow	-24.4	-13.0
	L-water	Distilled water	- 9.3	- 5.1
	M-3	Magnetite	3.0	1.7
	0-120	Quartz	9.5	4.8
	0-120	Quartz	9.6	4.9
	0-120	Quartz (fractionated)	13.2	6.4
	S-863	Quartz	17.6	9.1
Carbonaceous Chondrites				
Allende	Al-Mx	Matrix	1.3	- 3.1
	Al-Wr	Whole-rock	1.6	- 2.6
	Al- 3	Dark inclusion	3.3	- 1.3
	Al-17	White inclusion	-11.2	-14.1
	Al-15	White inclusion	-16.0	-18.2
	Al-15-1	Melilite	- 0.1	- 4.0
	Al-15-2	Melilite & spinel	- 6.7	-10.2
	Al-15-4	Pyroxene	-34.8	-35.7
	Al-15-5	Pyroxene & spinel	-39.0	-40.6
Murray	My-2	Olivine	- 6.9	- 9.7
Ornans	0- 4	Olivine	- 6.5	- 9.5
	0-10	Olivine	- 2.1	- 5.2
	0-12	Olivine	- 2.7	- 6.5
	0-13	Olivine	- 2.8	- 6.4
	0-14	Olivine	- 2.1	- 5.9
	0-15	Olivine	- 0.9	- 5.1
Vigarano	V-6	Olivine	- 4.4	- 7.9
Ordinary Chondrites				
Pantar	P-Wr	Whole rock	4.1	2.7
	P-A	Partial fluorination	7.3	4.6
	P-B	" "	6.5	4.3
	P-II-A	" "	5.4	3.8
	P-II-B	" "	5.6	3.9
	P-II-C	" "	7.8	4.9
Modoc	M-Wr	Whole rock	5.9	4.2

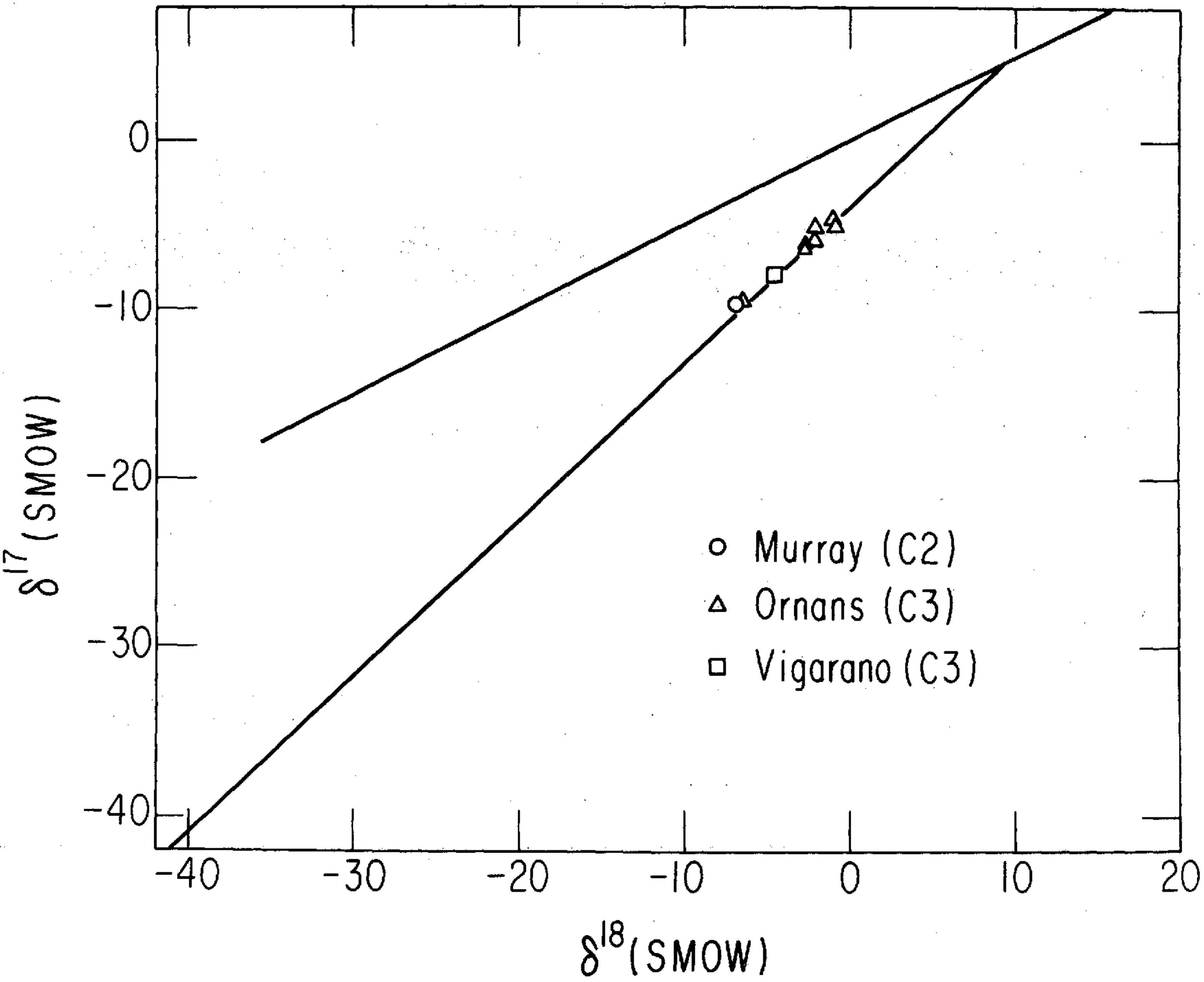
FIGURE CAPTIONS

Fig. 1 Oxygen isotopic compositions of various terrestrial materials, (squares) and of samples of the Allende meteorite (circles and triangles). The samples represented by circles are density separates from a single inclusion.

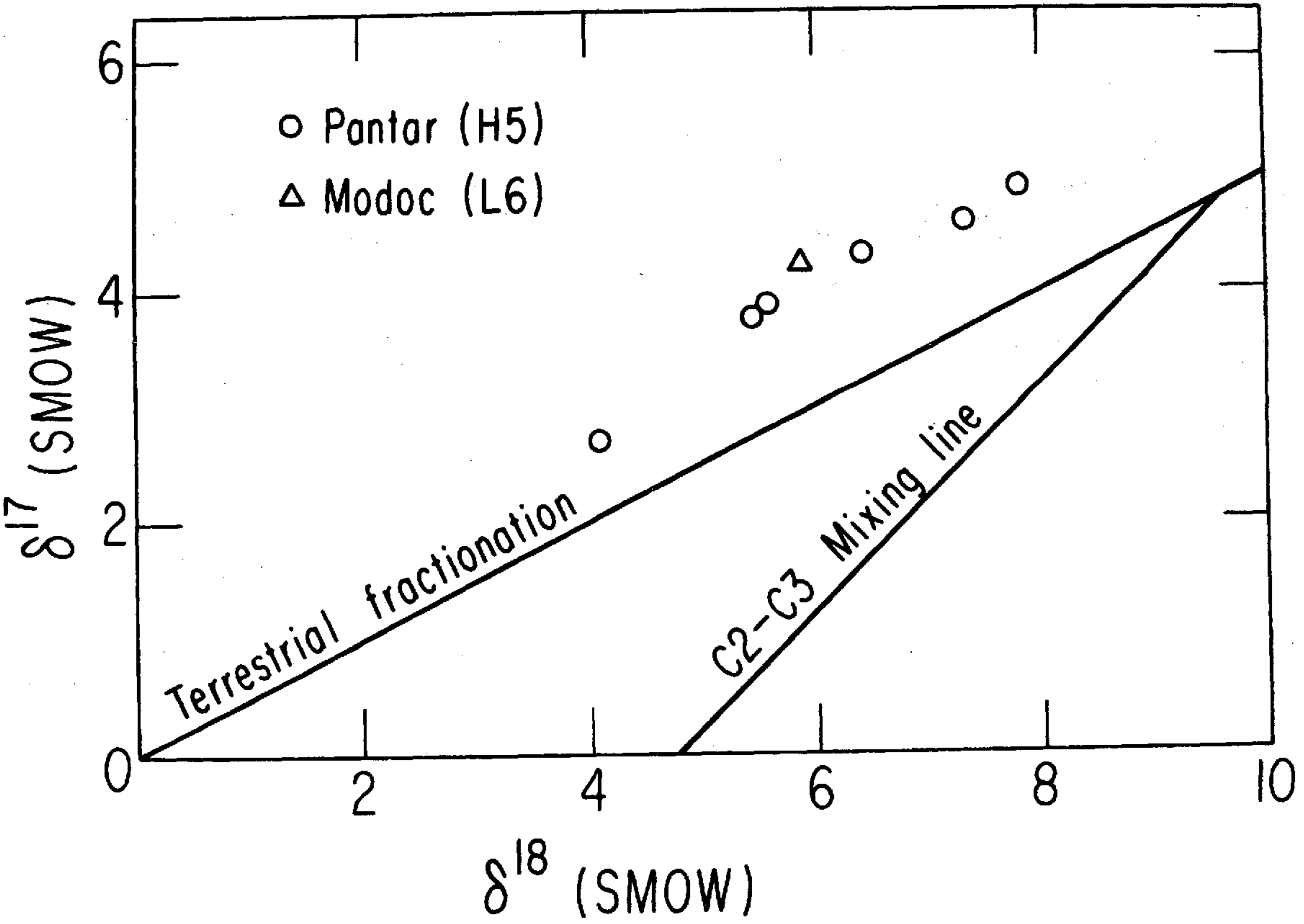
Fig. 2 Oxygen isotopic compositions of separated fractions of Murray, Ornans and Vigarano. The lines are taken from Fig. 1.

Fig. 3 Oxygen isotopic compositions of ordinary chondrites, Pantar and Modoc. The data from Pantar results from successive partial fluorinations; the Modoc result is a whole-rock analysis.


0
-10
-20
-30
-40
δ^{17} (SMOW)
-40
-30
-20
-10
0
10
20
δ^{18} (SMOW)



0
−10
−20
−30
−40
−40
−30
−20
−10
0
10
20
δ^{17} (SMOW)
δ^{18} (SMOW)
○ Murray (C2)
△ Ornans (C3)
□ Vigarano (C3)


○ Pantar (H5)
△ Modoc (L6)
Terrestrial fractionation
C2-C3 Mixing line
0
2
4
6
8
10
δ17 (SMOW)
δ18 (SMOW)