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IODINE-129
A STUDY OF ITS TRANSPORT IN THE ENVIRONMENT AND DISTRIBUTION IN BIOLOGICAL SYSTEMS

Final Progress Report

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APPENDIX I

IODINE-129 IN THYROIDS AND TELLURIUM ISOTOPES IN METEORITES BY NEUTRON ACTIVATION ANALYSIS

by

ROBERT VANGORDER BALLAD, 1945-

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ABSTRACT

A combination of neutron activation and mass spectrometry has been used to determine the concentration of fissiogenic ^{129}I and the value of the ^{129}I/^{127}I ratio in thyroids of man, cow, and deer from Missouri. Deer thyroids show an average value of $^{129}I/^{127}I = 1.8 \times 10^{-8}$ and an average concentration of 3×10^{-3} pCi ^{129}I per gram of thyroid (wet weight). Thyroids of cows and humans show successively lower values for the ^{129}I/^{127}I ratio and the ^{129}I content because their diets dilute fission-produced ^{129}I in the natural iodine cycle with mineral iodine. The results of analyses on a few thyroids from other geographic areas are also reported.

The isotopic compositions of tellurium, krypton, and xenon were determined in acid-resistant residues of the Allende meteorite. Neutron activation and γ-counting were used to determine the relative abundances of six tellurium isotopes, and mass spectrometry was used to determine the isotopic compositions of krypton and xenon in aliquots of the same residues. Nucleogenetic anomalies were observed in the isotopic compositions of these three elements. The presence of isotopically distinct components of tellurium, krypton, and xenon in these residues provides strong support for the suggestion that our solar system formed directly from the debris of a supernova.

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TABLE OF CONTENTS

Table of Contents (continued) Page

LIST OF ILLUSTRATIONS

Page

IODINE-129 IN MAN, COW AND DEER

NUCLEOGENETIC HETEROGENEITIES IN CHEMICAL AND ISOTOPIC ABUNDANCES OF THE ELEMENTS

ISOTOPES OF TELLURIUM, XENON AND KRYPTON IN ALLENDE RETAIN RECORD OF NUCLEOSYNTHESIS

LIST OF TABLES

INTRODUCTION

This dissertation describes the use of neutron activation for the analysis of two different types of samples. First is the determination of a rare iodine isotope, ^{129}I, in the presence of natural iodine, ^{127}I, in proportions which exist in the modern biosphere, $^{129}I/^{127}I \approx 10^{-8}$. Second is the determination of tellurium isotopes in meteorites.

Iodine-129 is a long-lived ($t_{\frac{1}{2}} = 1.7 \times 10^7$ yr) isotope of iodine. Although primordial ^{129}I is extinct, this isotope is produced with a 1% yield in the neutron-induced fission of ^{235}U and ^{239}Pu. Information on the distribution of ^{129}I in the environment has become increasingly important since the advent of the nuclear age. The long half-life of ^{129}I is responsible for both good and bad aspects of this isotope as an environmental contaminant from the use of nuclear weapons and reactors. A bad feature is that ^{129}I continues to accumulate as it is released to the environment. One good feature is that ^{129}I can be used as a long-term tracer in studies on the environmental pathways of fission-produced iodine, because the $^{129}I/^{127}I$ ratio is an indicator of dilution with natural iodine rather than decay of the fission product. Another advantage is the low specific activity and biological effects of ^{129}I.

The Rolla area is well suited for the study of environmental pathways of ^{129}I because the area is low in natural iodine and remote from any release point of ^{129}I. Thyroid glands were chosen for analysis because (i) the thyroid from one large animal contains sufficient iodine for ^{129}I analysis, (ii) the thyroid is seldomly consumed by man and is therefore readily donated for study, and (iii) iodine is easily extracted from thyroids.

Of the extraction methods tried in this study, base fusion, combustion in an oxygen furnace, and digestion with sulfuric acid followed by I_2 release on addition of H_2O_2, the latter method was found to be most reliable. Iodine from the thyroid, after cleanup, was precipitated as PdI_2. The precipitate was then sealed under vacuum, the PdI_2 decomposed by heat, and the iodine trapped on aluminum in a solid-CO_2 cooled quartz vial. The vial was then sealed under vacuum and heated to form Al_2I_6. The iodine was irradiated in this form, together with standards having known values of $^{129}I/^{127}I$ and triple neutron-capture monitors of natural ^{127}I. An irradiation time of 12 hours at a flux of 1×10^{14} n cm^{-2} sec^{-1} was found to be best for modern samples of iodine from thyroids. A flux of 5×10^{13} n cm^{-2} sec^{-1} was used for some samples which had lower ^{129}I concentrations than the modern thyroids.

The irradiation produces radioactive isotopes of iodine which decay to stable xenon isotopes, as shown below:

$$^{129}I(n,\gamma)^{130}I(\beta^-,\ t_{\frac{1}{2}} = 12.4\ hr)^{130}Xe$$

$$^{127}I(n,2n)^{126}I(\beta^-,\ t_{\frac{1}{2}} = 13\ d)^{126}Xe$$

$$^{127}\mathrm{I}(n,\gamma)^{128}\mathrm{I}(\beta^-,\ t_{\frac{1}{2}} = 25\ \mathrm{min})^{128}\mathrm{Xe}$$

$$^{127}\mathrm{I}(n,\gamma)^{128}\mathrm{I}(n,\gamma)^{129}\mathrm{I}(n,\gamma)^{130}\mathrm{I}(\beta^-,\ t_{\frac{1}{2}} = 12.4\ \mathrm{hr})^{130}\mathrm{Xe}$$

About 12 hours after the irradiation, the Al_2I_6 is dissolved and the solution flushed with N_2 to remove radiogenic ^{128}Xe. The iodine is reprecipitated as PdI_2, sealed under vacuum and the radiogenic ^{130}Xe and ^{126}Xe are allowed to accumulate several months. Then the xenon is released by vacuum decomposition of the PdI_2 and the concentration of radiogenic ^{130}Xe and ^{126}Xe determined by noble gas mass spectrometry. Values of the $^{129}I/^{127}I$ ratio are calculated from values of the $^{130}Xe/^{126}Xe$ ratio observed in the sample and monitors.

Results of this work, as shown in the first two papers, demonstrate that this is a reliable and sensitive method for determining the minute amounts of ^{129}I which exist in the local environment. The major disadvantage of the method is the amount of time required to accumulate radiogenic ^{126}Xe for analysis.

Acid-resistant residues of the Allende meteorite, shown by earlier studies to contain anomalous xenon, seemed to be good candidates for study in our search for other isotopically anomalous elements. Since neutron activation and gamma counting of the activated products were to be used for this study, it was necessary to select an element which met the following conditions: (i) The element should have an atomic number near that of xenon (Z = 54), (ii) the element should consist of several isotopes, including isotopes that were produced exclusively by the r-, p-, or s-processes of nucleosynthesis, (iii) these isotopes should form long-lived radioactive isotopes by the (n,γ) reaction and the activated products should emit identifiable gamma rays, (iv) the stable and the activated isotopes should be shielded from production by fission, (v) there should be negligible interference from other nuclear reactions that might produce the radioactive isotopes during the irradiation, e.g., (n,α) or (n,p) reactions, and (vi) the element should be one that can be quickly separated from all other elements in the postirradiation chemistry.

Element number 52, tellurium, meets almost all of the above conditions. It has eight stable isotopes of which six form easily detected (n,γ) products with half-lives of 30 hours to 120 days. Tellurium chemistry, precipitation from solution and separation from contaminating elements with holdback carriers of gold, platinum and selenium, is straightforward. The only unusual step used in this study was the sublimination of tellurium at reduced pressure as a final step in the cleanup procedure.

The results, as well as their implications for the formation of the solar system, are given in the last two papers. One unexpected result of the experiment was the discovery of excess ^{126}Te which might be the decay product of ^{126}Sn ($t_{\frac{1}{2}} \approx 10^5$ yr). If so, then this would add another short-lived isotope to the list of extinct radioactivities present in the early solar system and set stringent limits on the time interval between the end of element synthesis and the formation of meteoritic solids.

IODINE-129 IN THYROIDS OF GRAZING ANIMALS

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A combination of neutron activation and mass spectrometry has been used to determine the concentrations of fissiogenic ^{129}I and stable ^{127}I in thyroids of grazing animals and in mineral iodine. The $^{129}I/^{127}I$ ratios are lowest in mineral iodine and in a given area lower in cow thyroids than in deer thyroids. Near saturation levels of mineral iodine in commercial feeds and salt licks may account for differences in the ^{129}I levels of cows and deer. Values of the $^{129}I/^{127}I$ ratio in deer appear to vary inversely with the iodine concentration of the thyroid.

INTRODUCTION

Natural iodine has one stable isotope, ^{127}I, and one very long-lived isotope, ^{129}I, which decays with a half-life of 17×10^{6} years (Katcoff et al., 1951) to form stable ^{129}Xe. Studies of radiogenic ^{129}Xe in meteorites (Hohenberg et al., 1967) and in the Earth (Boulos and Manuel, 1971) indicate that natural iodine initially had an isotopic composition, $^{129}I/^{127}I \approx 10^{-4}$. A negligible fraction ($\approx 10^{-80}$) of the primordial ^{129}I remains today, but this isotope has been produced continually on Earth by spontaneous fission, by spallation reactions on Xe in the upper atmosphere, and by (n, γ) and $(n, 2n)$ reactions on ^{128}Te and ^{130}Te, respectively. Edwards (1962) estimates that the natural production of ^{129}I produced a steady state value of $^{129}I/^{127}I \approx 3 \times 10^{-14}$ in the hydrosphere, the atmosphere, and the biosphere. Measurements by Brauer et al. (1974a) show that pre-1936 human thyroids contained iodine with $^{129}I/^{127}I \leq 10^{-11}$, and studies by Srinivasan et al. (1971) indicate an isotopic composition, $2.2 \times 10^{-15} \leq {}^{129}I/^{127}I \leq 3.3 \times 10^{-15}$, for the iodine which formed the mineral iodyrite (AgI) about 50 million years ago.

Since the advent of the nuclear era, the inventory of ^{129}I on the Earth's surface has significantly increased as a result of induced fission in nuclear reactors and weapons. Owing to the 17×10^{6} year half-life of ^{129}I, it is not removed at a significant rate by decay and may continue to accumulate in the environment as a radioactive pollutant from nuclear technology. Bryant (1970), Russell and Hahn (1971) and Tadmor (1971) discuss public health aspects of ^{129}I, note the advantages of using the specific activity concept in evaluating the hazards of ^{129}I to man, and recommend upper limits on ^{129}I where $^{129}\mathrm{I}/^{127}\mathrm{I} \leq (0.9-5.4)10^{-2}$. Daly _et al_. (1974) and Matuszek _et al_. (1974) are critical of the specific activity concept and suggest that other factors indicate the need for more stringent limits on ^{129}I. All of these authors stress that the critical pathway to man for radioiodine discharged to the atmosphere is usually via grass-cow-milk, although fish (Matuszek _et al_., 1974) and fruits and vegetables (Thompson, 1967) could be more important for some segments of the population.

Compared to many fission products, there is little radiological health hazards associated with ^{129}I. This is one of several advantages considered by Studier et al. (1962) and Edwards (1962) in noting the potential utility of ^{129}I as an isotopic tracer. However, the slow decay rate of ^{129}I prohibits detection of minute quantities of this isotope by conventional radiation detectors, but Purkayastha and Martin (1956) demonstrated that a much higher sensitivity is available by neutron activation to produce ^{130}I. Studier _et al_. (1962) first employed neutron activation in studies of ^{129}I in environmental samples. The sensitivity of their procedure is illustrated by their results on analyzing leaves and finding an upper limit of 6×10^{-16} g ^{129}I per gram of leaf (Studier _et al_., 1962). This concentration of ^{129}I is about two orders of magnitude below the detection limited reported recently for a more elaborate procedure based on neutron activation and isotopic separation of the neutron capture product, ^{130}I, from other iodine isotopes (Rook *et al.*, 1975).

The results of activation analyses on ^{129}I in biological samples representing the 1944-1962 period indicate that values of the $^{129}\text{I}/^{127}\text{I}$ ratio in the biosphere of the United States increased from a value of $\approx 10^{-12}$ in 1944 to a peak value of $\approx 10^{-6}$ in 1950-1952, and then decreased to a value of $\approx 10^{-9}$ by 1962 (Keisch *et al.*, 1965). Iodine from human thyroids collected in Pittsburgh, Pennsylvania in 1962-1963 displayed a range of isotopic compositions with $10^{-10} \leq {}^{129}\text{I}/^{127}\text{I} \leq 10^{-8}$ (Koch and Keisch, 1964).

About two years ago we reported that modern thyroids contain iodine with $^{129}\text{I}/^{127}\text{I} \approx 10^{-8}$, in which neutron activation to produce ^{130}I was followed by mass spectrometric detection of the stable neutron-capture produce, ^{130}Xe (Boulos *et al.*, 1973). We have since learned of several detailed studies of ^{129}I in the vicinity of nuclear fuel reprocessing plants (Magno *et al.*, 1972; Brauer *et al.*, 1974a, b; Matuszek *et al.*, 1974; Gabay *et al.*, 1974; Daly *et al.*, 1974; Brauer and Ballou, 1975), which showed up to 3,700 pCi ^{129}I per gram of deer thyroid in 1970 (Matuszek *et al.*, 1974) and values for the $^{129}\text{I}/^{127}\text{I}$ ratio in cow thyroid as high as 1.6×10^{-3} (Magno *et al.*, 1972).

The present study on grazing animals was undertaken as part of our study aimed at determining the distribution and ecological pathways of ^{129}I in an area remote from sites where ^{129}I is being released. Brauer et al. (1974 a, b) and Brauer and Ballou (1975) recently published the results of a study underway since the 1950's on ^{129}I in various parts of the United States. As a cross-check on our methods, we included cow thyroids in our study from two of the areas covered by their investigation.

MATERIALS AND METHODS

A complete description of the procedure used in our laboratory to determine ^{129}I has been given earlier (Boulos et al., 1973). Briefly the method depends on mass spectrometric measurement of ^{128}Xe, ^{126}Xe, and ^{130}Xe produced as stable decay products of the following reactions:

$$^{127}I(n,\gamma)^{128}I \tag{1}$$

$$^{127}I(n,2n)^{126}I \tag{2}$$

$$^{129}I(n,\gamma)^{130}I \tag{3}$$

$$^{127}I(n,\gamma)^{128}I(n,\gamma)^{129}I(n,\gamma)^{130}I \tag{4}$$

The limits of detection are determined by the last reaction, triple neutron capture on ^{127}I.

Table 1 summarizes information on the samples used in this study. Six samples of iodine from the Chilian nitrate deposits were analyzed in order to determine the interference from triple neutron capture and thus measure the neutron capture cross section of ^{128}I. Studies by Claridge and Campbell (1968) indicate that these nitrate deposits have accumulated by precipitation from the atmosphere, and measurements by Brauer et al. (1974a) show iodine isotopes in these nitrate deposits with $^{129}\mathrm{I}/^{127}\mathrm{I} = (0.3\text{-}3)10^{-12}$.

The standard iodine monitor, UMR-IO-(129, 53) No. 4 was prepared in our laboratory from commercial ^{129}I purchased in 1969 from Nuclear Science, a division of the International Chemical and Nuclear Corporation (ICN).

Thyroid powder samples include hog thyroid powder purchased from the Sigma Chemical Company and a small sample of horse thyroid powder prepared at the Park Davis Research Laboratory in 1947 and supplied to us by Dr. L. Van Middlesworth. Fresh thyroids of cows were collected at local (Rolla) slaughter houses, those from San Francisco were purchased frozen (Pel Freeze Biologicals, Inc.), the one from Morris, Illinois was collected from a farm near the Dresden Power Reactor and supplied for our study by Dr. P. Tedeschi, and the bovine thyroids from Idaho Falls were collected from within a 60 mile radius of the Reactor Testing Station and supplied for our study by Dr. E. E. Eatinger. All of the deer thyroids were collected locally at check stations operated by the Missouri Conservation Commission during the 1974 deer season.

Iodine was extracted from the thyroids by digestion in H_2SO_4 (Rodgers and Poole, 1958; Borst Pauwels and Wasemael, 1962). The iodine was precipitated as $PdI_2 \cdot H_2O$ which was sealed in a quartz tube, decomposed by heat, and the free I_2 cold-trapped on Al powder. The latter was sealed under partial vacuum, heated to produce Al_2I_6, and irradiated for ≈ 22 hours at a flux of $\approx 6 \times 10^{14}$ n cm^{-2} sec^{-1}.

The samples were allowed to cool for 12 hours after the end of irradiation. Then the Al_2I_6 was dissolved in basic solution, degassed to remove radiogenic ^{128}Xe, and the iodine precipitated as $PdI_2 \cdot H_2O$. The latter was sealed under vacuum and allowed to collect radiogenic ^{130}Xe and ^{126}Xe for at least one month prior to analysis in a high-sensitivity mass spectrometer (Reynolds, 1956).

RESULTS AND DISCUSSION

The results of our analyses are shown in Table 2, where the samples are listed in the same order as in Table 1. Errors shown on the $^{129}I/^{127}I$ ratios represent one standard deviation (σ). Values given for the ratio were calculated from isotopic analyses for radiogenic ^{126}Xe and ^{130}Xe, the decay products of ^{126}I and ^{130}I which had been produced during the irradiation by reactions (2), (3), and (4). In our initial study using neutron activation and mass spectrometry to detect ^{129}I in thyroids (Boulos <u>et al.</u>, 1973), the iodine was irradiated at a flux $\approx 10^{14}$ n cm^{-2} sec^{-1}. This flux caused the production of ^{130}I by triple neutron capture, reaction (4), to equal that produced by simple neutron capture on ^{129}I, reaction (3), in a sample with $^{129}I/^{127}I \approx 10^{-10}$ (Boulos <u>et al.</u>, 1973). Since then the operating power of the University of Missouri Research Reactor has been increased such that iodine samples irradiated in the same position for the current study had an interference production of ^{130}I by reaction (4) equal to that produced by reaction (3) in samples with $^{129}\mathrm{I}/^{127}\mathrm{I} \approx 2 \times 10^{-8}$. In calculating the values of ^{129}I shown in Table 2, it was first necessary to correct the total amount of radiogenic ^{130}Xe in each sample for that resulting from triple neutron capture on ^{127}I. For this subtraction we assumed that iodine from the Chilian nitrate deposits contained $^{129}\mathrm{I}/^{127}\mathrm{I} << 10^{-8}$, as reported by Brauer et al. (1974a), and attributed all of the radiogenic ^{130}Xe in these samples to triple neutron capture on ^{127}I. A proportionate correction for triple-neutron-capture-produced ^{130}Xe was made on the other samples. This correction resulted in negative values for the $^{129}\mathrm{I}/^{127}\mathrm{I}$ ratio in a few of the samples shown in Table 2, but in all such cases the $^{129}\mathrm{I}/^{127}\mathrm{I}$ ratios are not distinguished from zero within the statistical error of the measurement. The effects of triple neutron capture will be used later to define the neutron capture cross-section of ^{128}I, but first we will discuss the results of our analyses of iodine in thyroids.

Our analyses provided no positive identification of ^{129}I in either sample of thyroid powder. The ^{129}I levels observed in bovine thyroids from San Francisco agree with the values reported in bovine thyroids collected from this area in 1971 (Boulos et al., 1973) and in bovine thyroids collected in California in 1965-1966 (Brauer et al., 1974a). Essentially the same $^{129}\mathrm{I}/^{127}\mathrm{I}$ ratio was observed in the sample of bovine thyroid collected from the area of the Dresden Power Reactor near Morris, Illinois. Our results on this sample seem to support the results of a recent study by Brauer and Ballou (1975) in which large excesses of ^{129}I were observed in the area of nuclear fuel reprocessing plants, but not in the area of the Dresden Power Reactor.

The highest levels of ^{129}I which we observed in bovine thyroids were in the two samples from Idaho. These results are also in general agreement with analyses by Brauer et al. (1974a), who report that bovine thyroids collected in Idaho in 1965-1966 contain iodine with $^{129}I/^{127}I = (0.33\text{-}52.8)10^{-8}$. Brauer et al. (1974a) report even higher levels of ^{129}I in bovine thyroids collected in the states of Washington and Oregon apparently due to ^{129}I released from the nuclear fuel separations plant at Hanford, Washington (Brauer et al., 1974b; Brauer and Ballou, 1975). In view of the general proximity of the states of Idaho, Oregon, and Washington, the ^{129}I which we observed in the Idaho bovine thyroids may be entirely due to the release of ^{129}I at Hanford and incidental to the Reactor Testing Station near Idaho Falls.

The iodine from thyroids collected in the general area of Rolla, Missouri show significantly higher levels of ^{129}I in deer than in cows. This is probably due to a larger fraction of mineral iodine in the diet of the latter. The natural iodine content of Missouri is low (Potter, 1930), such that mineral iodine in commercial feeds and salt licks may constitute a large part of the total iodine in the diets of cows in this area. In addition to diluting the more radioactive iodine ingested in grazing, Collard and Verly (1965) have shown that iodine-saturated thyroid glands will take up less iodine from a new feeding. Thus cows may be expected to excrete directly a larger fraction of iodine acquired in grazing than would deer.

The $^{129}I/^{127}I$ ratios in deer thyroids analyzed in this study generally vary inversely with the total iodine content of the thyroid. Thus, the total concentration of ^{129}I in the deer thyroids shows less variation than does the $^{129}I/^{127}I$ ratios. Differences in values for the $^{129}I/^{127}I$ ratios among deer and the lower values for $^{129}I/^{127}I$ ratios in cows from this area suggest that iodine-saturated thyroids may play an important role in shunting an appreciable fraction of the iodine which cows ingest in grazing, but that variations in the level of inorganic iodine in the diet of deer from this area act only in diluting the iodine which deer ingest in grazing. Additional samples are planned on deer and cow thyroids from this area in order to better define the effects of mineral iodine on the behavior of ^{129}I.

Table 3 summarizes the results this and other recent reports on ^{129}I in thyroids of grazing animals from different areas. These results are listed in order of slaughter dates. In general it appears that temporal variations in the levels of ^{129}I within the United States from 1962 to 1975 are minor compared to geographic variations. For example, iodine in the 1963 deer thyroids from Maryland (Brauer et al., 1974a) is not distinguishable from that formed in 1974 deer thyroids from Missouri. However, thyroids from deer in the vicinity of the nuclear fuel reprocessing plants near Hanford, Washington in 1964 and near West Valley, New York in 1970-1971 contained ≈ 3-5 orders of magnitude more ^{129}I than the deer thyroids analyzed in this study. The data shown in Table 3 also suggest that deer generally acquire higher levels of ^{129}I than cows.

As mentioned earlier, the interference level from triple neutron capture on ^{127}I was appreciable in this study and permit us to calculate a cross section for neutron capture on ^{128}I. Koch and Keisch (1964) indicated that this cross section is on the order of 100 barns, but Boulos et al. (1973) reported values of 3-9 barns. From the results of this study we obtain a cross section value for ^{128}I of 51 ± 12 barns for those samples irradiated at a flux of 6.23×10^{14} n cm^{-2} sec^{-1}, and a cross section value for ^{128}I of 110 ± 47 barns for samples irradiated at a flux of 5.59×10^{14} n cm^{-2} sec^{-1}. We also rechecked the data of Boulos et al. (1973) and found that their results yield an average value for the ^{128}I cross section of 44 barns for samples irradiated at a flux of $\approx 1 \times 10^{14}$ n cm^{-2} sec^{-1}.

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Table 1. Iodine samples analyzed in this study

Source of Iodine	Sample Number	Date Isolated from Source	Geographic Location	Number of Animals in Sample
Mineral				
Nitrate Deposits	172-1-A	1973	Chile	0
Nitrate Deposits	172-1-B	1973	Chile	0
Nitrate Deposits	172-1-C	1973	Chile	0
Nitrate Deposits	172-1-D	1973	Chile	0
Nitrate Deposits	172-1-E	1973	Chile	0
Nitrate Deposits	172-1-F	1973	Chile	0
Standard				
Prepared from Commercial Fission Products	UMR-IO-(129, 53) No. 4	1968	USA	0
Thyroid				
Horse, Commercial Powder	51-1	1947	USA	Not Known
Hog, Commercial Powder	4M	1972	USA	Not Known
Cow	SF-1222	1973	San Francisco, CA	30
Cow	SF-419	1973	San Francisco, CA	5
Cow	M-2	1973	Morris, IL	1
Cow	R-9	1974	Rolla, MO	1
Cow	R-3	1974	Rolla, MO	1
Cow	R-2	1974	Rolla, MO	1
Cow	IF-421	1973	Idaho Falls, ID	4
Cow	IF-211	1973	Idaho Falls, ID	1
Deer	D-53	1974	Rolla, MO	1
Deer	D-69	1974	Rolla, MO	1
Deer	D-71	1974	Rolla, MO	1
Deer	D-82	1974	Rolla, MO	1
Deer	D-89	1974	Rolla, MO	1

Table 2. Results of analyses

Sample Number	Weight of Iodine (mg)	^{129}I/^{127}I Atom Ratio (10^{-8})	^{127}I Concn. (mg/g thyroid)	^{129}I Concn. (p Ci/g thyroid)
<u>Mineral</u>				
172-1-A	20.4	0.04 ± 0.21	----	---------
172-1-B	10.8	-0.44 ± 0.91	----	---------
172-1-C	5.2	0.65 ± 1.54	----	---------
172-0-A	142	0.07 ± 0.18	----	---------
172-0-B	74	-0.03 ± 0.04	----	---------
172-0-C	109	0.03 ± 0.15	----	---------
<u>Standard</u>				
UMR-IO-(129, 53) No. 4	10	$\equiv 1.000$	----	---------
<u>Thyroid</u>				
51-1	4.0	0.04 ± 0.40	----	---------
4M	8.4	-0.02 ± 0.27	----	---------
SF-1222	20.6	0.77 ± 0.30	1.08	1.4×10^{-3}
SF-419	33.5	0.76 ± 0.30	1.12	1.4×10^{-3}
M-2	5.2	0.79 ± 0.33	0.28	0.4×10^{-3}
R-9	11.5	-0.42 ± 0.60	1.69	$\leq 0.6 \times 10^{-3}$
R-3	9.4	-0.78 ± 0.91	1.75	$\leq 0.4 \times 10^{-3}$
R-2	17.2	0.82 ± 0.67	1.87	2.5×10^{-3}
IF-421	19.0	3.7 ± 1.3	1.46	9.0×10^{-3}
IF-211	9.6	30.2 ± 10.5	0.46	23×10^{-3}
D-53	17.2	3.2 ± 1.1	2.17	12×10^{-3}
D-71	6.6	3.4 ± 1.2	2.08	12×10^{-3}
D-69	4.4	5.4 ± 2.0	1.57	14×10^{-3}
D-82	2.9	5.1 ± 1.9	0.61	5.2×10^{-3}
D-89	1.7	18.6 ± 9.9	0.56	17×10^{-3}

Table 3. Iodine-129 in thyroids of grazing animals from different areas

Animal	Location and Date	$^{129}I/^{127}I$ Atom Ratio (10^{-8})	^{129}I Activity (p Ci/g thyroid)	Reference
Sheep	Hanford Reservation, WA, 1962	86-905	0.140-0.905	Brauer et al. (1974a)
Deer	Maryland, 1963	6.5	0.006-0.040	Brauer et al. (1974a)
Elk	Wyoming, 1963	18.0-18.5	0.097-0.248	Brauer et al. (1974a)
Deer	Blue Mountains, OR, 1963	406-672	0.276-0.525	Brauer et al. (1974a)
Deer (fetus)	Blue Mountains, OR, 1963	604-879	0.142-0.681	Brauer et al. (1974a)
Sheep	Nashville, TN, 1964	1.38	-----------	Boulos et al. (1973)
Sheep	Wyoming, 1964	8.7-11.2	0.017-0.049	Brauer et al. (1974a)
Antelope	Wyoming, 1964	18.4-19.4	0.020-0.140	Brauer et al. (1974a)
Deer (fetus)	Hanford Reservation, WA, 1964	3,100-4,400	1.61-2.37	Brauer et al. (1974a)
Deer	Hanford Reservation, WA, 1964	-----------	6.36-10.8	Brauer et al. (1974a)
Sheep	Nashville, TN, 1966	0.68	-----------	Boulos et al. (1973)
Sheep	Nashville, TN, 1966-1967	0.89	-----------	Boulos et al. (1973)
Deer	NFS, West Valley, NY, 1968	-----	< 80-120	Matuszek et al. (1974)
Deer	NFS, West Valley, NY, 1969	-----	< 80-1,200	Matuszek et al. (1974)
Cow	Boston, MA, 1970	-----	< 0.017-0.035	Matuszek et al. (1974)
Cow	Boston, MA, 1970	< 22-42	< 0.014-0.017	Magno et al. (1972)
Deer	NFS, West Valley, NY, 1970	-----	< 80-3,700	Matuszek et al. (1974)
Cow	San Francisco, CA, 1971	1.05	--------	Boulos et al. (1973)
Cow	Boston, MA, 1971	11-35	0.016-0.040	Mango et al. (1972)
Cow	West Valley, NY, 1971	-----	3.5-179	Matuszek et al. (1974)
Cow	West Valley, NY, 1971	1,050-162,000	3.6-190	Magno et al. (1972)
Deer	NFS, West Valley, NY, 1971	-----	< 80-1,400	Matuszek et al. (1974)
Cow	West Valley, NY, 1972	-----	0.2-12.7	Matuszek et al. (1974)
Deer	NFS, West Valley, NY, 1972	-----	27-360	Matuszek et al. (1974)
Cow	Morris, IL, 1973	0.8	0.0004	This work
Cow	San Francisco, CA, 1973	0.8	0.0014	This work
Cow	Idaho Falls, ID, 1973	3.7-30.2	0.009-0.023	This work
Deer	NFS, West Valley, NY, 1973	--------	20-65	Matuszek et al. (1974)
Cow	Rolla, MO, 1974	< 0.13-0.82	< 0.0004-0.0025	This work
Deer	Rolla, MO, 1974	3.2-18.6	0.005-0.017	This work

IODINE-129 IN MAN, COW AND DEER

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Abstract—Concentrations of ^{129}I and values of the ^{129}I/^{127}I ratio have been measured in over forty individual thyroids of human, cow and deer from Missouri. Deer thyroids show an average value of ^{129}I/^{127}I = 1.8 x 10^{-8} and ^{129}I concentrations of 3 x 10^{-3} pCi per g of thyroid (wet weight). Thyroids of cow and human show successively lower values for the ^{129}I/^{127}I ratio and for the ^{129}I content due to dilution of ^{129}I from the natural geochemical cycle with mineral iodine in their diets. Analyses on a few thyroids from other areas are also reported.

Natural iodine consisted of about 99.99% ^{127}I and 0.01% ^{129}I when this element was incorporated into meteorites (Ho67) and the Earth (Bo71) about 4.6 x 10^9 years ago (Ma75). The half-life of ^{129}I is short, $t_{\frac{1}{2}} = 1.57 \times 10^7$ yrs (Nu72), on the geologic time scale, so that the supply of primordial ^{129}I became extinct in the early history of the solar system. However, it was estimated (Ed62) that the production of ^{129}I on Earth by fission of actinides, by neutron-induced reactions on tellurium and by spallation reactions on Xe in the upper atmosphere would produce a steady state value of $^{129}I/^{127}I \approx 3 \times 10^{-14}$ in the hydrosphere, the atmosphere and the biosphere. This estimate has been confirmed (He77) by recent analyses on radiogenic ^{129}Xe in the natural mineral iodyrite (AgI).

Since the advent of the nuclear era, ^{129}I levels in the biosphere of the United States increased to a peak value of $^{129}I/^{127}I \approx 10^{-6}$ in the 1950's (Ke65) and then declined to the modern value of $^{129}I/^{127}I \approx 10^{-8}$ (Bo73, Ba76). The latter represents an average biospheric value for the United States; values of $^{129}I/^{127}I \approx 10^{-5} - 10^{-3}$ have been reported in mammalian thyroids from the vicinity of nuclear fuel reprocessing plants (Ma72, Br74, Ma74).

Unlike other radioactive iodine isotopes produced by fission, environmental ^{129}I does not decay at an appreciable rate relative to the time scales of recorded history. The long half-life of ^{129}I causes this to accumulate in the environment as a pollutant of nuclear technology. This same property makes ^{129}I a useful tracer isotope in determining the environmental transport of this radionuclide into an area and its passage into the food chain (Ed62, St62). Measurements on the $^{129}I/^{127}I$ ratio are particularly useful in identifying various stages in the transport of ^{129}I, since the $^{129}I/^{127}I$ ratio is diluted with natural iodine at each stage. This property has been used to trace ^{129}I in the vicinity of nuclear fuel reprocessing plants (Ma72, Br75).

Previous reports from this laboratory (Ba76, Bo73) have described the method for detecting trace amounts of ^{129}I and the $^{129}I/^{127}I$ ratio in environmental samples which are remote from ^{129}I sources. This study gives the results of additional analyses on ^{129}I in mammalian thyroids from Missouri. Since this area has been shown to be low in natural iodine (Po30), man-made additives of mineral iodine to the natural food supply may account for a large part of the iodine ingested by man and domesticated animals. To show the mixing of fission-produced ^{129}I with other iodine in this area, analyses were performed on iodine from deer, cows and man.

Deers are browsers, eating leaves, twigs, shoots, and in season acorns and cultivated crops of the area such as corn. This diet would avoid man-introduced iodine, except in isolated cases where deer might be exposed to mineral iodine in artificial salt licks. Cattle also graze from a natural diet, but iodine acquired in this manner is diluted with mineral iodine in salt blocks and other food supplements. Humans receive some natural iodine in sea foods, fruits and leafy vegetables (Th67), but the bulk of iodine ingested by man in this goitrous area (Po30) is probably from additives of mineral iodine in salt and prepared foods.

Animal thyroids were chosen for analyses because they contain about 60% of the iodine in mammals (Ce69), the iodine is easily extracted from this organ, and the long biological half-life (about 180 days in humans) of iodine in thyroids should yield time-averaged ^{129}I levels (Ce69).

MATERIALS AND METHODS

The analytical procedure is based on a combination of neutron activation and high sensitivity mass spectrometry. During the irradiation, ^{126}I is produced via the $^{127}I(n,2n)$ reaction and ^{130}I is produced via the $^{129}I(n,\gamma)$ reaction. Following the irradiation, short-lived activities, such as ^{128}I from the $^{127}I(n,\gamma)$ reaction, are allowed to decay for 12 hours. The irradiated iodine is then precipitated as PdI_2 and sealed under vacuum in glass vials. Radiogenic ^{126}Xe and ^{130}Xe are extracted from the vials and analyzed in the mass spectrometer (Re56) several months after the irradiation. Irradiation monitors, made by dilutions on Standard Reference Material 4949 of the National Bureau of Standards (NBS-SRM 4949), were analyzed by the same procedure. Details of the analytical procedure have been presented earlier (Bo73, Ba76).

RESULTS AND DISCUSSION

The results of our analyses are presented in Table 1. The errors listed in Table 1 are one standard deviation (σ) and are the quadratic sum of statistical errors in mass spectrometric measurements on samples, irradiation monitors and calibration Xe spikes. All of the deer thyroids analyzed in this study were provided by cooperative hunters at the Phelps County deer checking station during a two week period of the 1974 deer season in Missouri. Each analysis represents an individual thyroid for the samples collected in Missouri. The horse and pork thyroids were commercially prepared thyroid powder representing many animals. A few of the analyses on cow thyroids from other areas were performed on composite samples from several animals. The number in parenthesis in the first column indicates the number of individuals in these composite samples. For comparison purposes given earlier (Ba76), analyses were performed on thyroids of cattle from a few other states. The mean values shown in Table 1 were computed only for thyroids from a common area.

In a previous paper (Ba76) we reported the results of analyses on thyroids identified by the underlined sample numbers. A recent cross-check of the ^{129}I monitor used in that study against ^{129}I in NBS-SRM 4949 indicated that our earlier monitor contained appreciably less ^{129}I than assumed (Ba76). Corrected values for ^{129}I in these samples, computed relative to monitors of NBS-SRM 4949 irradiated in the same position, are included in Table 1.

The number of analyses on bovine thyroids from the Rolla area are too small for a reasonable statistical analysis. However, there appears to be no discernible pattern of ^{129}I levels in cattle from different locations, except for the prominent excess of ^{129}I in one of the bovine samples, IF 211, from the area of the Idaho Falls Reactor Testing Station. All of the analyses shown in Table 1 for cattle thyroids are included in Fig. 1, except for the two suspect Idaho samples from the close proximity of the Reactor Testing Station. The distribution pattern seems to be consistent with that expected when iodine acquired in grazing is diluted by iodine in feed supplements.

Values observed for the ^{129}I/^{127}I ratios in human thyroids all fall into the lowest grouping. This might be expected for diets where iodine from foods containing higher ^{129}I/^{127}I ratios, such as milk, fruits and vegetables, constitutes only a small fraction of the total iodine injested.

The trends seen in ^{129}I/^{127}I values in Fig. 1 are also reflected in Table 1 by the mean values of the ^{129}I/^{127}I ratios for these three types of thyroids. The average concentrations of ^{129}I, expressed as pCi per gram of thyroid, show a similar trend.

The amount of ^{129}I present in the horse thyroid powder of 1947 could not be detected within the experimental uncertainty of our measurement. The ^{129}I/^{127}I ratio in powdered thyroid tissue of hogs is similar to that observed in human thyroids. Similarities in the ^{129}I/^{127}I ratios found in thyroids of man and domesticated hogs probably indicate that most of the iodine in their diets is from food additives.

The yearly dose rate to humans from ^{129}I in an area remote from a source of this radionuclide can be estimated by assuming a steady state ^{129}I/^{127}I ratio. From the values obtained in this study, we estimate that an average 16 g thyroid contains a total of 4.4×10^{-3} pCi of ^{129}I. Book (Bo77) gives a conversion factor of 0.06 mrem yr^{-1} pCi^{-1} for an adult thyroid. This yields a yearly dose of 2.6×10^{-4} mrem/yr to the adult thyroid. This dose is insignificant relative to the dose from background radiation.

CONCLUSIONS

The results of this study show the dilution of radioactive ^{129}I by mineral iodine which has been added to the diets of man and domesticated animals. The results of analyses currently in progress on ^{129}I in rain, streams, insects and wild vegetation may elucidate the mode of transport of ^{129}I into an area remote from sources of fission-produced ^{129}I.

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Table 1. Iodine-129 in thyroid glands

Sample	Date Collected	Geographic Location	$^{129}I/^{127}I$ Atom Ratio (10^{-9})	^{127}I Conc (mg/g thyroid)	^{129}I Conc (pCi/g thyroid)
Deer					
89	Nov 1974	Rolla, MO	67 ± 27	0.56	67 x 10^{-4}
144	Nov 1974	Rolla, MO	32.8 ± 2.6	0.40	24 x 10^{-4}
138	Nov 1974	Rolla, MO	24.5 ± 2.5	0.55	24 x 10^{-4}
84	Nov 1974	Rolla, MO	23.6 ± 0.7	0.67	28 x 10^{-4}
137	Nov 1974	Rolla, MO	20.6 ± 1.0	1.07	40 x 10^{-4}
130	Nov 1974	Rolla, MO	20.2 ± 0.9	0.66	24 x 10^{-4}
75	Nov 1974	Rolla, MO	20.2 ± 0.5	0.71	26 x 10^{-4}
69	Nov 1974	Rolla, MO	19.7 ± 1.9	1.57	56 x 10^{-4}
82	Nov 1974	Rolla, MO	18.3 ± 3.1	0.61	20 x 10^{-4}
93	Nov 1974	Rolla, MO	18.1 ± 0.5	1.51	49 x 10^{-4}
129	Nov 1974	Rolla, MO	18.0 ± 0.5	0.62	20 x 10^{-4}
67	Nov 1974	Rolla, MO	16.6 ± 1.0	0.57	16 x 10^{-4}
140	Nov 1974	Rolla, MO	15.6 ± 0.9	1.78	50 x 10^{-4}
170	Nov 1974	Rolla, MO	15.4 ± 0.6	0.82	23 x 10^{-4}
81	Nov 1974	Rolla, MO	15.1 ± 0.5	1.85	50 x 10^{-4}
85	Nov 1974	Rolla, MO	12.6 ± 0.5	1.72	40 x 10^{-4}
142	Nov 1974	Rolla, MO	12.5 ± 2.7	0.75	17 x 10^{-4}
71	Nov 1974	Rolla, MO	12.4 ± 1.1	2.08	46 x 10^{-4}
91	Nov 1974	Rolla, MO	12.2 ± 0.6	1.01	22 x 10^{-4}
53	Nov 1974	Rolla, MO	12.0 ± 0.7	2.17	47 x 10^{-4}
79	Nov 1974	Rolla, MO	12.1 ± 4.7	0.56	12 x 10^{-4}
156	Nov 1974	Rolla, MO	11.9 ± 0.9	0.45	10 x 10^{-4}
125	Nov 1974	Rolla, MO	9.2 ± 3.4	2.02	33 x 10^{-4}
167	Nov 1974	Rolla, MO	8.5 ± 0.9	0.37	6 x 10^{-4}
124	Nov 1974	Rolla, MO	6.3 ± 1.1	0.39	4 x 10^{-4}
78	Nov 1974	Rolla, MO	5.0 ± 0.5	0.90	8 x 10^{-4}
Mean[a]		Rolla, MO	17.7	1.01	29 x 10^{-4}

Table 1. Iodine-129 in thyroid glands (cont.)

Sample	Date Collected	Geographic Location	$^{129}I/^{127}I$ Atom Ratio (10^{-9})	^{127}I Conc (mg/g thyroid)	^{129}I Conc (pCi/g thyroid)
Cow					
IF-211	July 1973	Idaho Falls, ID	108 ± 1.1	0.46	89×10^{-4}
R-33	Oct 1974	Rolla, MO	15.1 ± 1.4	0.27	7×10^{-4}
R-180	June 1975	Rolla, MO	6.1 ± 0.5	0.95	10×10^{-4}
SF-1301	July 1973	San Francisco, CA	5.6 ± 0.4	0.31	3×10^{-4}
IF-421 (4)	July 1973	Idaho Falls, ID	4.7 ± 0.5	1.46	12×10^{-4}
R-2	Sept 1974	Rolla, MO	3.7 ± 2.6	1.87	12×10^{-4}
SF-1222 (30)	July 1973	San Francisco, CA	3.1 ± 0.5	1.08	6×10^{-4}
SF-419 (5)	July 1973	San Francisco, CA	3.1 ± 0.4	1.12	6×10^{-4}
R-20	Sept 1974	Rolla, MO	3.1 ± 0.7	0.42	2×10^{-4}
M-2	May 1973	Morris, IL	2.8 ± 0.6	0.28	1×10^{-4}
SF-1221 (30)	July 1973	San Francisco, CA	2.8 ± 0.3	1.08	5×10^{-4}
R-178	June 1975	Rolla, MO	1.6 ± 0.5	0.95	3×10^{-4}
R-3	Sept 1974	Rolla, MO	-2.1 ± 3.5	1.75	$\leq 4 \times 10^{-4}$
R-9	Oct 1974	Rolla, MO	-0.8 ± 2.5	1.69	$\leq 5 \times 10^{-4}$
Mean[a]		Rolla, MO	5.9	0.89	7×10^{-4}
Human					
60	Oct 1974	St. Louis, MO	4.6 ± 0.2	0.74	6×10^{-4}
61	Oct 1974	St. Louis, MO	3.3 ± 0.9	0.42	2×10^{-4}
58	Oct 1974	St. Louis, MO	2.8 ± 0.5	0.28	1×10^{-4}
59	Oct 1974	St. Louis, MO	2.1 ± 0.5	2.19	8×10^{-4}
64	Oct 1974	St. Louis, MO	1.6 ± 0.6	0.38	1×10^{-4}
62	Nov 1974	St. Louis, MO	1.4 ± 0.6	0.79	2×10^{-4}
55	Oct 1974	St. Louis, MO	1.1 ± 0.7	0.55	1×10^{-4}
65	Oct 1974	St. Louis, MO	1.1 ± 0.8	0.58	1×10^{-4}
Mean[a]		St. Louis, MO	2.3	0.74	3×10^{-4}

Table 1. Iodine-129 in thyroid glands (cont.)

Sample	Date Collected	Geographic Location	$^{129}I/^{127}I$ Atom Ratio (10^{-9})	^{127}I Conc (mg/g thyroid)	^{129}I Conc (pCi/g thyroid)
Pork[b]					
54-3 (many)	1972	USA	1.7 ± 0.6	----	--------
54-4 (many)	1972	USA	1.6 ± 0.2	----	--------
54-5 (many)	1972	USA	2.1 ± 0.3	----	--------
4M (many)	1972	USA	-0.1 ± 1.6	----	--------
Mean[c]	1972		1.8	----	--------
Horse[b]					
51-1 (many)	1947	USA	0.1 ± 2.1	----	--------

[a] Mean values are only for thyroids from Missouri. Negative values of ^{129}I in samples R-3 and R-9 of cow thyroids were not included in this average.

[b] Commerically prepared powered thyroids.

[c] Negative value of ^{129}I in sample M4 of pork thyroid powder was not included in this average.

Figure 1

Distribution of $^{129}I/^{127}I$ ratios in thyroids of human (hatched bar), cow (solid bars) and deer (open bars) from Missouri. Values of $^{129}I/^{127}I$ are grouped by increments of 0.5×10^{-8}.

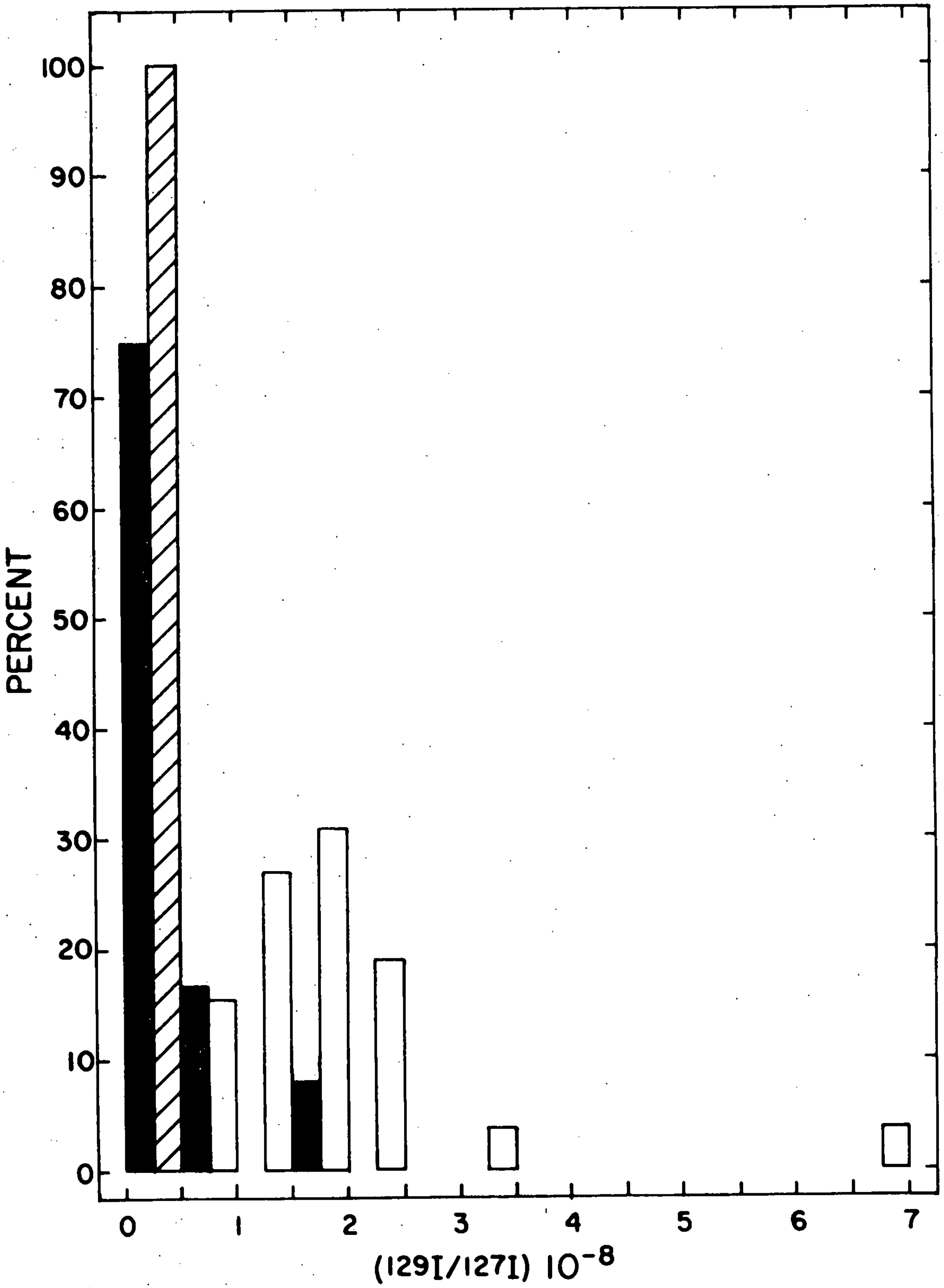

Figure 1

NUCLEOGENETIC HETEROGENEITIES IN CHEMICAL AND ISOTOPIC ABUNDANCES OF THE ELEMENTS

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Six years ago Manuel <u>et al</u>. (1972) suggested that the isotopically anomalous Xe in carbonaceous chondrites might represent a mixture of r- and p-products that had been added to the solar system from a nearby supernova. Later Manuel and Sabu (1975,1977) noted that meteoritic abundances of He and Ne correlate with abundances of isotopically anomalous Ar, Kr, and Xe but that neither was trapped in meteorite phases containing normal Ar, Kr, and Xe. They concluded that these observations in noble gases, chemical gradients in the planetary system, and the low flux of solar neutrinos are evidence that the solar system formed directly from debris of a single supernova and that (i) the sun grew on the supernova's core, (ii) iron meteorites and the iron-rich cores of terrestrial planets formed primarily from elements synthesized in the central stellar region, (iii) the internal structures of terrestrial planets were generated by inhomogeneous accretion as material from other stellar regions was drawn toward the sun, and (iv) the outer planets and the carbon-rich portions of carbonaceous chondrites formed from elements of the outer stellar layers. A test of their conclusion, based on the isotopic compositions of noble gases in the metal phase of iron meteorites, was reported by Hennecke and Manuel (1977). The results of another test, based on the isotopic compositions of Te in Allende's carbon (the host phase of anomalous Xe), is presented here.

We employed neutron activation and γ-ray spectrometry to determine the abundances of six stable Te isotopes and mass spectrometry to measure Xe isotopes. The isotopic anomaly patterns which we observed in Te and Xe extracted from Allende's carbon are shown in Fig. 1. Values on the vertical axis are

$$g^i_m = ({}^iX/{}^mX)_{\text{sample}}/({}^iXe/{}^mX)_{\text{standard}},$$

where ${}^iX$ and ${}^mX$ are stable isotopes of element X. The Xe spectra are normalized to ${}^{130}Xe$, and AVCC is used as the standard. The Te spectra are normalized to ${}^{124}Te$, and terrestrial Te is used as the standard. The isotopic composition of AVCC Te is not known, but De Laeter et al. (1978) report terrestrial abundances of Te isotopes in eight bulk meteorite samples.

The empty circles in Fig. 1 indicate that ${}^{129}Xe$ and ${}^{126}Te$ abundances may contain radiogenic products from in situ decay of precursor isotopes, ${}^{129}I$ ($t_{\frac{1}{2}} = 17 \times 10^6$ yr) and ${}^{126}Sn$ ($t_{\frac{1}{2}} \approx 10^5$ yr), respectively. The nonradiogenic isotopes of Te and Xe in Allende's carbon, depicted by filled circles, produce remarkably similar isotopic anomaly patterns. Because our analyses yielded an atomic ratio of Te:Xe $\approx 10^4$ in Allende's carbon and revealed large enrichments of neutron-rich and neutron-poor isotopes of both elements, we conclude that the anomalous Xe is nucleogenetic, rather than the product of carbonaceous-chondrite-fission, CCF (Podosek et al., 1971), or superheavy-element-fission (Lewis et al., 1975).

Additional evidence for nucleogenetic heterogeneities in elemental, as well as isotopic, compositions of the primitive nebula is shown in Fig. 2. Here values of the $^{136}Xe/^{132}Xe$ ratio in residues of Allende (Lewis et al., 1975) and Murchison (Srinivasan et al., 1977) are plotted against two elemental ratios, $^{4}He/^{132}Xe$ and $^{20}Ne/^{132}Xe$, and one isotopic ratio, $^{124}Xe/^{132}Xe$. We did not determine He and Ne, but isotopic ratios of Xe in the carbon- and spinel-rich fractions of our Allende residues (Fig. 2c) lie along the isotope correlation line derived from these earlier analyses. Isotopes of Xe in the spinel-rich fraction have an opposite anomaly pattern to that in carbon, i.e., Xe in the spinels is enriched in the middle isotopes, and values of $^{124}Xe/^{132}Xe$ and $^{126}Xe/^{132}Xe$ are essentially atmospheric. We also observed a slight enrichment (2.4 ± 0.6%) of the $^{124}Te/^{128}Te$ ratio in Allende's spinels relative to terrestrial Te. An earlier observation of enriched s-products in Xe from Murchison (Srinivasan and Anders, 1978) was attributed to alien nucleogenetic material from red giant stars. Since the isotopic compositions of Xe in the carbon- and spinel-rich fractions of Allende lie on the same correlation line in Fig. 2c, we suggest that these fractions trapped remnants of different nucleosynthetic processes which collectively produced the isotopes of bulk meteoritic Xe and other high-Z elements of the solar system. Isotopic compositions of Xe which lie along the vertical line in Fig. 2c, such as Xe in air and in iron meteorites, may be understood as mixtures of the s-enriched Xe of spinels with heavy Xe isotopes that are generated by the r-process that occurs near the supernova core (Trimble, 1975).

Correlations between abundances of He and Ne with abundances of isotopically anomalous Xe in residues of carbonaceous chondrites (Fig. 2a, b) yield zero values for He and Ne at a value of $^{136}Xe/^{132}Xe$ which is intermediate to values in bulk carbonaceous chondrites, in which $^{136}Xe/^{132}Xe = 0.321$ (Eugster et al., 1967), and in bulk ureilites, in which $^{136}Xe/^{132}Xe = 0.313$ (Phinney, 1971; Wilkening and Marti, 1976). The He and Ne have normal planetary isotopic compositions (Reynolds et al., 1978), they are trapped in a carbon matrix with a normal value of $^{12}C/^{13}C$ (Takahashi et al., 1976), but they are accompanied by Xe which is enriched in isotopes generated by the r- and p-processes that occur in supernova envelopes.

We conclude that the primitive nebula retained large-scale elemental and isotopic heterogeneities from the stellar processes which synthesized our elements. Injections of alien nucleosynthetic material into an otherwise homogeneous nebula, on the interstellar dust grains (Clayton, 1976), from a nearby supernova (Cameron and Truran, 1977), red giant star (Srinivasan and Anders, 1978), etc., cannot explain (i) the presence of isotopically anomalous Xe and Te in isotopically normal carbon, (ii) the association of anomalous Xe, Kr, and Ar with normal He and Ne in one noble gas component, (iii) the absence of He and Ne in another component, or (iv) the fact that both Xe enriched in s-products and Xe enriched in r- and p-products fit a single isotopic correlation line. These observations indicate that planetary solids did not condense from a homogeneous nebula, as assumed in many condensation calculations, but from the heterogeneous debris of a single supernova.

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Figure 1

Isotopes of Te and Xe in Allende's carbon display similar anomaly patterns. Empty circles represent isotopes which might contain radiogenic components from decay of long-lived r-products, filled circles represent other isotopes, and triangles represent standard isotopic compositions.

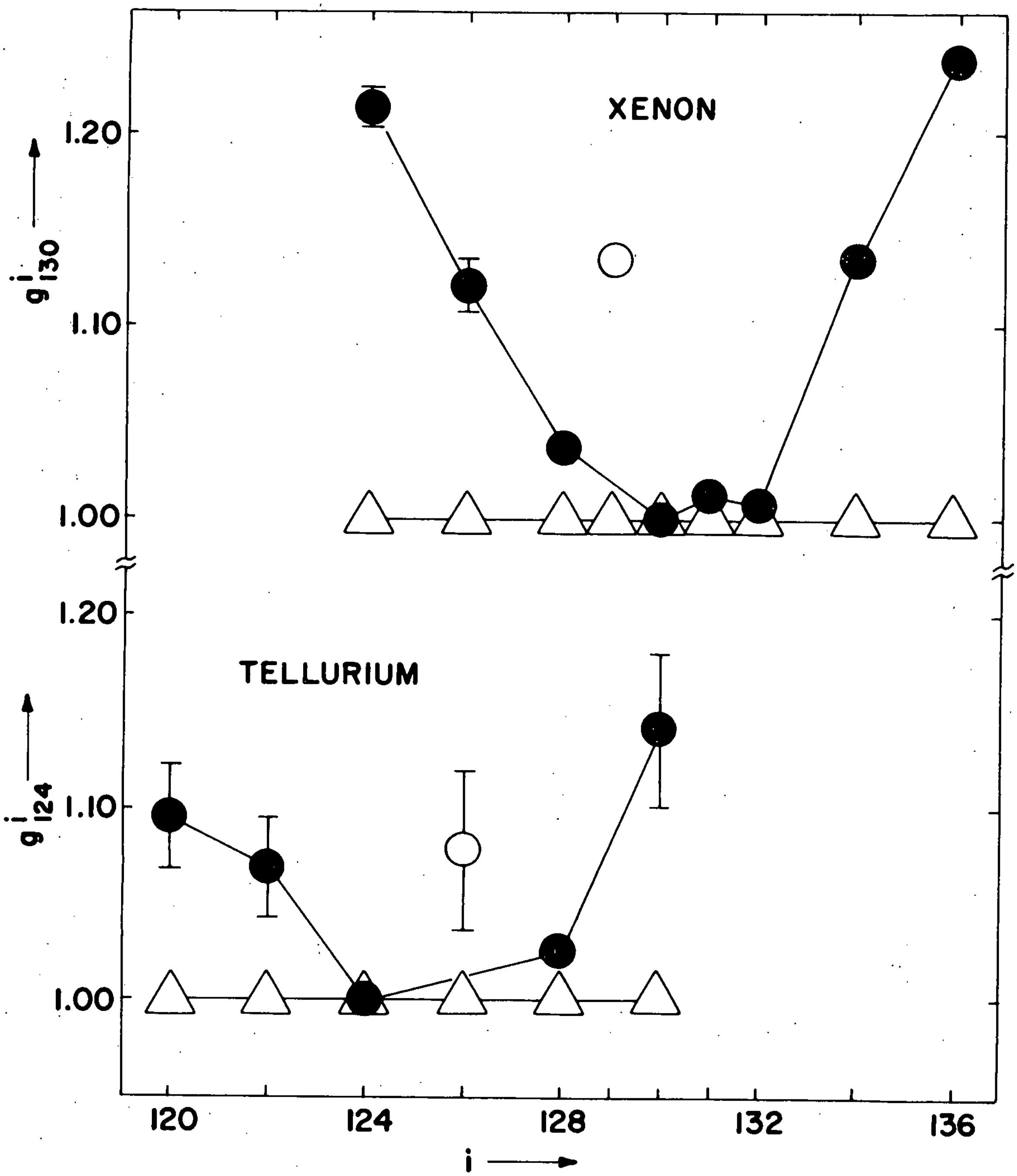
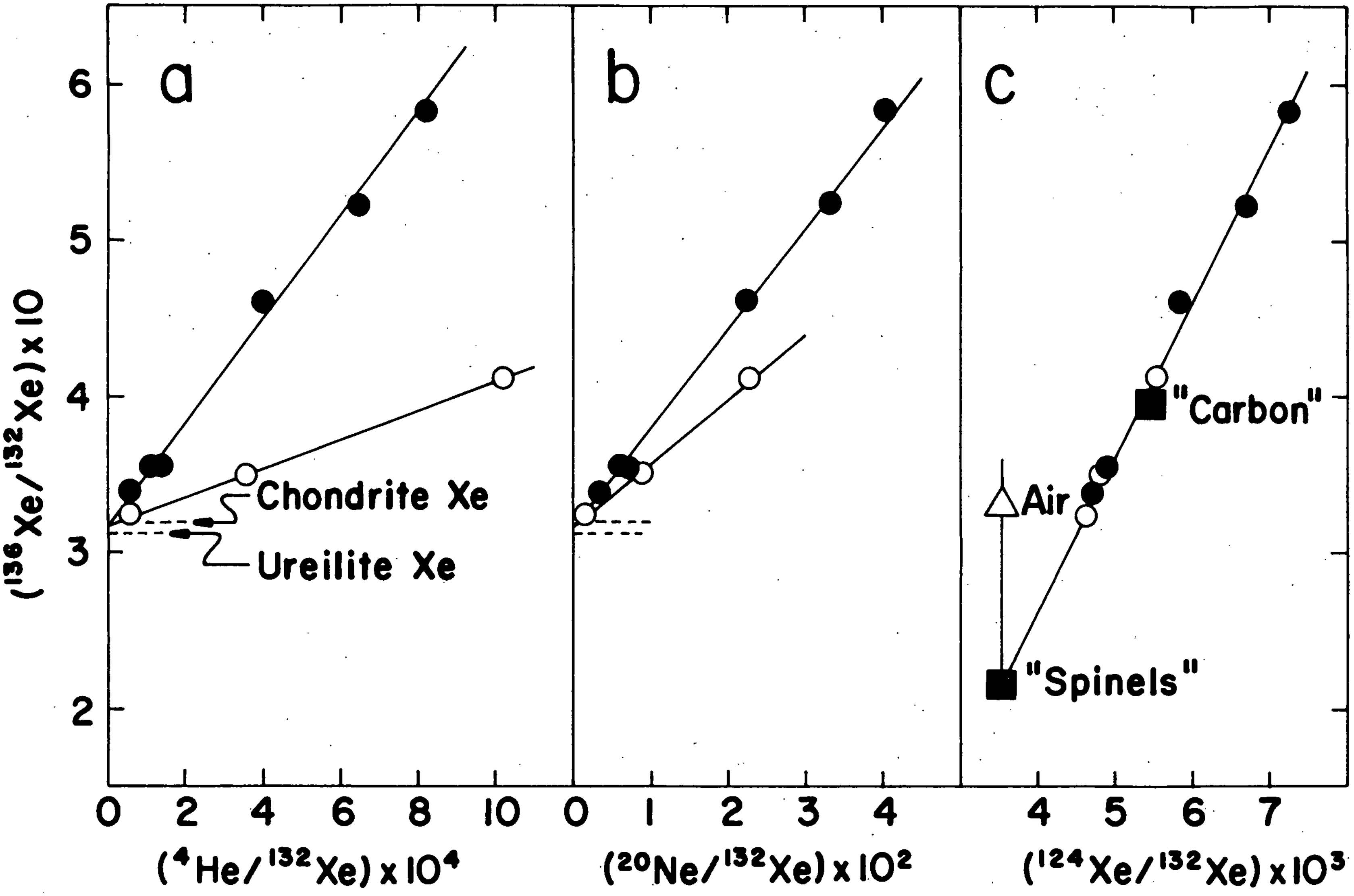

XENON
TELLURIUM
g^i_{130}
g^i_{124}
1.20
1.10
1.00
1.20
1.10
1.00
120
124
128
132
136
i

Figure 1

Figure 2

Values of the $^{136}Xe/^{132}Xe$ ratio in residues of carbonaceous chondrites correlate with elemental ratios of low-Z:high-Z noble gases and with other isotopic ratios of Ar, Kr, and Xe. Filled circles represent gases in Allende residues (Lewis et al., 1975), empty circles represent gases in Murchison residues (Srinivasan et al., 1977) and squares represent gases in our Allende residues. Correlations in Fig. 2a and Fig. 2b indicate that He and Ne in the primitive nebula were associated with Xe having values of $^{136}Xe/^{132}Xe > 0.6$, but that no He or Ne was associated with Xe having values of $^{136}Xe/^{132}Xe < 0.3$. Correlations in Fig. 2c show that Xe in carbon is enriched in r- and p-products and that Xe in spinels is enriched in s-products, but these lie along a common correlation line and demonstrate incomplete mixing of products of the different nucleosynthesis reactions that produced the isotopes of bulk Xe in carbonaceous chondrites.

Figure 2


a
b
c
(^{136}Xe/^{132}Xe) x 10
6
5
4
3
2
Chondrite Xe
Ureilite Xe
Air
"Carbon"
"Spinels"
0
2
4
6
8
10
(^{4}He/^{132}Xe) x 10^{4}
0
1
2
3
4
(^{20}Ne/^{132}Xe) x 10^{2}
4
5
6
7
(^{124}Xe/^{132}Xe) x 10^{3}

ISOTOPES OF TELLURIUM, XENON AND KRYPTON IN ALLENDE RETAIN RECORD OF NUCLEOSYNTHESIS

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Various mixtures of r-, p- and s-nucleosynthesis products have been observed in the isotopes of tellurium, xenon and krypton which we extracted from mineral separates of the Allende meteorite. The presence of several isotopically distinct components in these high Z elements and their close association with isotopically normal components of low Z elements provide strong support for the suggestion that our solar system formed directly from the debris of a single precursor star, approximately concentric with the present sun.

In 1972 it was noted that the enrichment of heavy xenon isotopes in carbonaceous chondrites is always accompanied by an enrichment of the light, fission-shielded isotopes. It was suggested that this anomalous xenon might represent a mixture of r- and p-products that had been added to the solar system from a nearby supernova[1]. Previously the excess heavy isotopes were widely regarded as fission products, designated CCF as an acronym for carbonaceous chondrite fission[2]. More recent reports of nucleogenetic anomalies in the isotopic compositions of several other elements in meteorites[3-10] and identification[11,12] of the decay product of extinct ^{26}Al have resulted in three different models of element synthesis shortly before condensation: (i) The anomalous elements and short-lived isotopes might have been generated in remote nucleosynthesis events, such as supernovae[13,14] or red giant stars[15], and then trapped in grains which were subsequently incorporated into the early solar nebula. In such a case the short-lived isotopes themselves would not necessarily survive until the grains entered the solar nebula, but the more refractory dust grains might survive conditions in the nebula and be incorporated into meteorites with alien nucleosynthesis products and the decay products of short-lived isotopes. (ii) The anomalous elements and short-lived isotopes might have been generated in a nearby supernova[16]. If the shock wave from this supernova caused collapse of the presolar nebula, then the nucleosynthesis event is directly related to the birth of the solar system and occurred about 4.6×10^9 yrs ago.

(iii) The solar system might have formed directly from the debris of a single supernova[17,19]. In this case the anomalous elements and short-lived isotopes were made by the same nucleosynthesis events which produced other elements in the solar system, and the present solar system retains the chemical characteristics of the supernova: The sun accumulated on the supernova core, iron meteorites and cores of terrestrial planets were formed primarily from elements synthesized in the hot stellar interior, and the outer planets and the carbon phase of chondrites were condensed from elements of the cooler outer layers, the only region which contained low-Z elements.

Two recent reviews[20,21] suggest that model (ii), a nearby supernova, has gained favor as a reasonable working hypothesis. However the results of analyses on noble gases trapped in the metal phase of iron meteorites[22] and in gas-rich xenoliths[23] from the Earth's mantle have been interpreted as supporting model (iii). Other tests were suggested for model (iii), including its prediction that the acid-resistant carbon which contains anomalous xenon should also contain isotopically anomalous components of other high-Z elements[22]. Subsequently, Arden[6] has reported large isotopic anomalies of uranium in acid-resistant minerals of several chondrites, but it was uncertain whether these were generated by stellar nuclear reactions or by chemical fractionation of the precursor elements of ^{235}U and ^{238}U. The present study was therefore undertaken to test the prediction of model (iii) at tellurium (Z = 52), an element with an atomic number near that of xenon (Z = 54) and with eight natural isotopes that are only inefficiently produced by fission.

Srinivasan followed procedures developed elsewhere[24-27] to isolate the following acid-resistant fractions from a crushed sample of bulk Allende weighing 11.66 g. (a) Residue A: Etching bulk Allende in HCl and HF acids for a six week period produced residues weighing 0.53% of the bulk sample. Previous studies[24-27] on Allende residues suggest that carbon, chromite, spinel and an ill-defined mineral Q are the main constituents of residue A. (b) Residue B: Etching residue A in concentrated HNO_3 dissolved Q (ref. 26,27), leaving a residue weighing 0.42% of the bulk Allende sample. (c) Residue C: Refluxing residue B in a mixture of concentrated acids, H_3PO_4 and H_2SO_4, for two hours dissolved[28] the chromite and spinels (and perhaps part of the carbon) to produce residue C, weighing 0.19% of the bulk sample. (d) Dissolution of C: Refluxing residue C for one hour in a mixture of concentrated acids, H_3PO_4 and H_2SO_4, plus CrO_3, dissolved residue C.

Nucleosynthetic products in xenon and krypton

A high sensitivity noble gas mass spectrometer was used to determine the isotopic compositions of xenon and krypton released by stepwise heating of a 3.1 mg aliquot of residue B, labeled B-1, and a 1.7 mg aliquot of residue C, labeled C-2. Procedures for extraction and analysis of noble gases have been described previously[22]. The isotopic compositions of xenon and krypton in these residues are presented in Tables 1 and 2, respectively. The xenon in both residues displays the now familiar pattern of enrichment in the light and heavy isotopes.

The part (chromite, spinel and perhaps some carbon) of residue B which dissolved (54% by weight) in producing residue C is referred to collectively as spinels in Tables 1 and 2. The concentrations and isotopic structures of gases in these spinels, as shown in Tables 1 and 2, were calculated from differences in the isotopic compositions and total gas contents of residues B and C. Superficially the isotopic composition of xenon and krypton in Allende spinels is similar to that reported recently in the high temperature fractions of Murchison residues which were etched in alkaline oxidants to remove organic polymers[15]. It was reported that x-ray diffraction of the severely etched Murchison residues showed mainly chromite and minor spinels—minerals which probably constitute the bulk of our Allende spinels. As was observed in the high temperature fractions of these Murchison residues[15], the xenon in Allende spinels is enriched in ^{130}Xe and depleted in ^{136}Xe, and the krypton in these spinels seems to be enriched in ^{82}Kr and depleted in ^{83}Kr. However, the isotopic composition of Kr in our spinels cannot be distinguished from bulk krypton in carbonaceous chondrites or in air, within the limits of experimental error. A detailed examination of the xenon spectrum in Allende's spinels is instructive in evaluating the model proposed to explain the high temperature xenon in Murchison's spinel—s-products trapped in presolar dust grains that were ejected from red giant stars[15].

$$g^{i}_{132} = ({}^{i}\mathrm{Xe}/{}^{132}\mathrm{Xe})_{\mathrm{sample}}/({}^{i}\mathrm{Xe}/{}^{132}\mathrm{Xe})_{\mathrm{AVCC}} \qquad (1)$$

Since the <u>in situ</u> decay product of ^{129}I may comprise part of the ^{129}Xe abundance, this isotope is not used to define the general anomaly pattern of isotopes trapped in Allende's spinels and carbon. Abundances of the other xenon isotopes in carbon and spinels display opposite anomaly patterns. We suggest that these mineral fractions trapped different components of AVCC xenon, i.e., carbon is enriched in r- and p-products and spinels are enriched in s-products of the nucleosynthesis reactions which collectively produced the isotopes of bulk xenon in carbonaceous chondrites. It seems unlikely that the carbon and spinel fractions are both presolar grains, or that either carbon or the spinels forming in the primitive nebula were selectively exposed to alien xenon from nearby supernovae or red giant stars. There are other indications that noble gases in Allende do not represent alien nucleosynthesis products. Allende's carbon contains helium and neon with normal planetary isotopic compositions[29], and the carbon itself has a normal value of ${}^{12}C/{}^{13}C$ (ref. 30). The ^{82}Kr/^{130}Xe ratio in Allende's spinels is 2.6, which is close to the value of ${}^{82}\mathrm{Kr}/{}^{130}\mathrm{Xe} = 1.8$ in bulk Allende[26] and about a factor of 10 higher than the value that Srinivasan and Anders[15] report for this ratio in the gas component from red giant stars.

Variations in the abundance of the lightest and heaviest xenon isotopes in gas released by stepwise heating of Allende's acid-etched residues are shown graphically in Fig. 2. Xenon isotopes released from the carbon (C-2) and spinel/carbon (B-1) fractions define different correlation lines. These do not agree with the correlation shown by Lewis et al.[26,27] for other etched residues of Allende (dashed line in Fig. 2), nor do they pass through the composition of AVCC or "trapped" (ref. 26) xenon. Since ^{132}Xe may be produced by both r- and s-processes[31], it is preferable to normalize the xenon spectra to a pure s-product, ^{130}Xe.

As shown in Fig. 3a, this method of normalization permits us to compare trends in isotopic ratios with trends expected from the addition of pure s-, r- and p-nucleosynthesis products to AVCC xenon. It can be seen in Fig. 3b that xenon isotopic ratios from stepwise heating of our two Allende residues do not follow the trend reported earlier[26,27] (dashed line in Fig. 3a), but it can be seen in Fig. 3c that ratios for the total xenon in these residues seem to do so. Isotopic ratios of the total xenon in other etched residues of Allende[26,27] also lie along the dashed line shown in Fig. 3a, as expected by mixing only two components. But isotopic ratios of xenon released by stepwise heating of individual residues from Allende and Murchison, as shown in Figs. 3d, 3e and 3f, cannot be explained by a simple binary mixture. Kuroda[32] also notes that a simple binary mixture fails to explain isotopic ratios of xenon released by stepwise heating of bulk carbonaceous chondrites. Although the spectra in Fig. 3 confirm the positive correlation between enrichments of light and heavy xenon isotopes[1], and the spectra in Fig. 1 suggest that bulk AVCC xenon may be presented as a mixture of one s-enriched component with one s-depleted component, differences in the slopes of the correlation lines in Fig. 3 indicate that the r- and p-products of chondritic xenon were not completely mixed before trapping. Since the r- and p-processes occur in the supernova explosion[31], to explain these xenon spectra as a consequence of injections from a nearby supernova[1,16] would require the preservation of heterogeneities within the supernova material itself as the alien xenon was trasferred into the solar nebula and then trapped on a matrix of isotopically normal carbon.

Variations in the isotopic composition of krypton (Table 2) provide additional evidence for nucleogenetic effects in the residues of Allende. Since the anomalies in krypton are relatively small and others[15,33] have recently discussed the evidence for s- and r-products in krypton extracted from acid-etched minerals of carbonaceous chondrites, a detailed discussion on the krypton anomalies shown in Table 2 is not warranted.

Nucleosynthetic products in tellurium

If Allende's carbon itself formed in the outer layers of a supernova, then other heavy elements trapped in the carbon would be expected to show enrichments of r- and p-products similar to those observed in xenon. To test this, the abundance of six stable isotopes, ^{i}Te at i = 120, 122, 124, 126, 128 and 130, was determined in Allende's residues by neutron activation and γ-ray spectrometry on the (n, γ) product, $^{i+1}$Te. For these analyses a 32 mg aliquot of residue B, labeled B-3, was irradiated for 95 hours at a flux of 6×10^{14} neutrons cm^{-2} sec^{-1}. Monitors of tellurium, uranium, osmium and ruthenium were irradiated with the sample. Carriers of tellurium, iodine (to carry fissiogenic ^{133}I), osmium, and ruthenium were added to the irradiated sample during etching to isolate the activation products from fractions of the residues rich in spinels and carbon, respectively. Standard radiochemical procedures were used to recover tellurium[34-38] and iodine[39], and to recover and separate osmium and ruthenium[35,40-45]. The only special reagent, potassium chlormilexathiocyanate, was prepared as described by Morgan[45] and used in the precipitation of osmium and ruthenium. Attempts to measure the isotopic compositions of osmium and ruthenium were abandoned after our results showed that these elements are highly enriched on the residues, apparently as a result of their insolubility in the etching acids[46], and display no large isotopic anomalies.

A Ge(Li) detector and a thin-crystal, intrinsic Ge detector were used with a 1024 channel analyzer to determine the activity of each tellurium isotope relative to the activity of ^{129}Te in the samples and in the monitors. The latter were prepared from purified TeO_2, lot 766061, Fisher Scientific Co., Fair Lawn, New Jersey. Possible interferences which might produce $^{i+1}$Te from neutron-induced reactions on other elements in the samples are (i) neutron-induced fission of uranium to produce ^{131}Te and ^{129}Te, (ii) the (n, p) reaction on iodine and the (n, α) reactions on xenon, and (iii) the (n, γ) on ^{124}Sn followed by two successive β-decays. Our results indicate ^{128}Te concentration of 16 ppm in the carbon and 21 ppm in spinels, corresponding to total tellurium content of 51 ppm and 67 ppm, respectively, if all other isotopes were present in the proportion observed for terrestrial tellurium. The activity of fission-produced ^{133}I and ^{132}Te in each residue and in the uranium monitor indicate that interference from neutron-induced fission of uranium produces 1.1% of the ^{131}Te activity and 0.5% of the ^{129}Te activity in the irradiated carbon, and fission interference produces 0.9% of the ^{131}Te and 0.4% of ^{129}Te activities in the irradiated spinels. The results of our tellurium and xenon analyses (Table 1) yield atomic ratios of Te/Xe $\approx 5 \times 10^4$ in carbon and and Te/Xe $\approx 1.5 \times 10^5$ in spinels. From the amounts of radiogenic ^{129}Xe in these residues (Table 1) and an assumed value of $^{129}I/^{127}I = 10^{-4}$ for trapped iodine[47], we calculate atomic ratios of Te/I $\approx$ 130 in carbon and Te/I $\approx$ 480 in the spinels. The ^{127}I (n, p) reaction and the

$^{124-134}$Xe (n, α) reactions have Q values of about 0.1 Mev and 2.5-6.3 Mev, and they are inhibited by Coulomb barriers of about 8 Mev and 14 Mev, respectively. Thus, each of the interference reactions listed under item (ii) requires neutron energies above 8 Mev. All of the relevant cross-sections for (n,p) and (n, α) reactions are not known, but typically these are only a few millibarns for 14 Mev neutrons on target elements with Z $\approx$ 50-60 (ref. 48). In the position where our samples were irradiated, the flux of neutrons above 8 Mev is about 1×10^{-3} of the thermal flux. From the atomic ratios given above and assuming cross-sections of 1000 millibarns for each (n,p) and (n, α) reaction, we estimate upper limits for interference on any tellurium isotope to be 0.03% from the (n,p) reaction and 1×10^{-4}% from the most favorable (n, α) reaction. Concentrations of tin, the target element for interference from above item (iii), were not measured in our Allende residues. In other Allende residues prepared in a similar manner, the highest concentration of germanium, a neighbor of tin in the Group IV A elements, is reported[46] to be 37 ppm. If our Residue B (carbon + spinels) contained 37 ppm germanium, we calculate lower limits on the atomic ratios of Te/Sn $\geq$ 11 in carbon and Te/Sn $\geq$ 18 in the spinels, assuming a solar value for the Sn/Ge ratio[49] and assuming that the mineral fraction being considered, carbon or spinels, each contain all of the germanium of Residue B. The ^{125}Sn produced by neutron-capture on natural ^{124}Sn must undergo two β-decays before becoming an interference at ^{125}Te. An intermediate decay product,

^{125}Sb ($t_{\frac{1}{2}}$ = 2.73 yr), acts to shield ^{125}Te from most of the ^{125}Sn decays, and we calculate an upper limit of 0.1% for the interference at ^{125}Te from the ^{125}Sn (n, γ, β, β) reaction.

The results of counting data accumulated from February 5, 1978 to May 28, 1978 are shown in Table 3, corrected for the fission interference discussed above. It should be stressed that we measured only relative abundances of tellurium isotopes in the monitors and in the samples. The maximum variation that Smithers and Krouse[50] observed in isotope ratios in six natural tellurides and two commercial tellurium salts corresponds to a fractionation of only 0.05% per mass unit, variations which could not be detected by the techniques used in this study. We therefore assume that the tellurium in our monitors have normal values for isotope ratios, and that the activity of each isotope, $^{i+1}$Te, is proportional to the abundance of the stable (n, γ) target isotope, ^{i}Te. Thus, the activities shown in Table 3 can be expressed in an analogous manner to that given for xenon isotopes in equation (1).

$$g^{i}_{128} = (^{i}\mathrm{Te}/^{128}\mathrm{Te})_{\mathrm{sample}}/(^{i}\mathrm{Te}/^{128}\mathrm{Te})_{\mathrm{standard}} \qquad (2)$$

The results of our analyses on tellurium isotopes in Allende residues are shown graphically in Fig. 4. Since ^{126}Te may contain a radiogenic decay product from in situ decay of a precursor isotope produced in the r-process, ^{126}Sn ($t_{\frac{1}{2}} \approx 10^{5}$ yr) (ref. 51), the abundance of ^{126}Te is not used to define the general anomaly pattern of tellurium isotopes trapped in these residues. No data are available on the isotopic composition of tellurium in average carbonaceous chondrites, AVCC tellurium. Therefore, the patterns of isotopic anomalies shown in Fig. 4a (carbon) and Fig. 4b (spinels), where the isotopic ratios are normalized to the terrestrial tellurium standard, are not analogous to the patterns shown in Fig. 1 for xenon in these same residues. The xenon spectra in Fig. 1 are normalized to an AVCC xenon standard.

In spite of differences in reference standards used in Figs. 1 and 4, the following observations may be significant: (i) The nonradiogenic isotopes of tellurium and xenon in Allende's carbon, as shown in Fig. 1 and Fig. 4a, produce remarkably similar anomaly patterns. Both elements are enriched in the heaviest isotopes, ^{130}Te and ^{136}Xe, and in the lightest isotopes, ^{120}Te and ^{124}Xe. These isotopes are produced on a very short time scale by the r- and p-processes in supernova explosions[31, 52]. (ii) The spinel-rich fraction of these minerals (Figs. 1 and 4b) is enriched in the intermediate mass isotopes of each element, ^{130}Xe and ^{124}Te. These isotopes are generated over long periods of stellar evolution by the s-process[31,15]. (iii) The existence of isotopically anomalous tellurium in Allende cannot be explained by a single AVCC-type tellurium, since the isotopic spectrum of tellurium in carbon is obviously different from that in the spinels. (iv) The spinel-rich fraction of Allende residues is enriched in ^{130}Te (Fig. 4b) but not in ^{136}Xe (Fig. 1). This apparent discrepancy between the abundances of r-products in tellurium and xenon may indicate that etching the Allende residues has separated the r- and p-products of tellurium, just as stepwise gas extraction separated the r- and p-products of xenon (Fig. 3). Or this may be due to differences in the reference standards for the xenon and tellurium spectra, i.e., it is possible that AVCC tellurium is systematically enriched in ^{130}Te relative to terrestrial tellurium.

Nucleogenetic heterogeneities in element abundances

In using the results of this study to evaluate the merits of models suggested for element synthesis shortly before condensation[13-19], it is imperative to consider the constraints imposed by earlier analyses[26] on noble gases in Allende's residues. These measurements revealed elemental abundances of helium and neon which correlate linearly with abundances of the r-products in xenon[17-19], as shown in Fig. 5, and the correlations yield zero values of helium and neon at ^{136}Xe/^{132}Xe = 0.32 ± 0.01. This value of ^{136}Xe/^{132}Xe contains the range of isotopic ratios for planetary xenon in air, carbonaceous chondrites and ureilites. It is now widely accepted that variations of the ^{136}Xe/^{132}Xe ratio in carbonaceous chondrites may be nucleogenetic[1,5,9,13-23,29,33], and that stellar evolution selectively depletes low-Z elements from the hotter stellar interior[31]. It can be concluded that normal, planetary xenon was synthesized in the hot stellar interior and that the inertness of noble gases made it possible for these elements to preserve a record of <u>both elemental and isotopic heterogeneities</u> which existed prior to condensation. Most other elements formed compounds which were chemically segregated into different minerals of planetary solids. Small variations in the isotopic compositions of normal planetary xenon, as seen in bulk ureilites[53,54], carbonaceous chondrites[55], and air[56], are identified in Fig. 5 at values of $^{136}Xe/^{132}Xe = 0.313, 0.322$ and 0.330, respectively. Correlation coefficients between the elemental and isotopic ratios shown in Fig. 5 are between 99.2% and 99.9%. It has been reported[26] that a plot of $^{124}Xe/^{130}Xe$ against $^{136}Xe/^{130}Xe$ for xenon in these same residues gave a correlation coefficient of 98%.

We did not determine helium and neon in our study but have used the trends shown in Fig. 5 in interpreting our results, since the results of other studies on noble gases in residues of Allende[27,33,57] and Murchison[58] confirm these correlations. These correlations are also seen in bulk noble gases of meteorites. Most carbonaceous chondrites contain helium and neon, and stepwise heating[1] or chemical etching experiments[26] on carbonaceous chondrites reveal the presence of an anomalous xenon component with $^{136}Xe/^{132}Xe > 0.32$. However, Marti and Wilkening[59] report that the stepwise release of gases from the Kenna ureilite revealed the presence of a single xenon component with $^{136}Xe/^{132}Xe = 0.312$, but they found no evidence of trapped helium or neon although the ureilite contained large amounts of heavy noble gases.

Conclusions

The presence of isotopically anomalous tellurium in Allende provides additional evidence for the suggestion that isotopically anomalous components of xenon in carbonaceous chondrites are nucleogenetic[1, 5, 9, 13-23, 29, 33] rather than fission products[2, 26, 27, 46, 58]. The following observations are difficult to explain by alien grains carrying nucleosynthetic elements from supernovae[13, 14] or red giant stars[15]: (i) The carbon grains have a normal value of $^{12}C/^{13}C$ (ref. 30), (ii) the carbon contains anomalous xenon, krypton and tellurium and normal helium and neon[29], (iii) isotopic ratios of xenon enriched in s-products lie on the correlation line defined by isotopic ratios of xenon enriched in r- and p-products (Fig. 3a, c), and (iv) there was no helium or neon in the primitive noble gas component which contained normal, planetary xenon (Fig. 5 and ref. 59). Trapping of alien nucleosynthetic elements from a nearby supernova[1, 16] onto a matrix of isotopically normal material can explain observations (i) and (ii), but not observations (iii) and (iv). The observation that r- and p-products from the supernova event are not mixed (Fig. 2, Fig. 3b, d-f, Fig. 4) also requires that the injected material remain heterogeneous as it was transferred from the supernova to the solar system. Even if this occurred, it seems highly unlikely that the abundance pattern of xenon isotopes from supernovae[1,16] and red giant stars[15] would be mirror images and that these two alien forms of xenon would be fortuitously trapped in meteorites in which the bulk xenon isotopes display an intermediate abundance pattern (Fig. 1).

The isotopically anomalous components of tellurium, xenon and krypton observed in this study can be understood as remnants of the different nucleosynthesis processes which collectively produced the chemical elements of the solar system. The opposite anomaly patterns of xenon (Fig. 1), the incompletely mixed r- and p-products in xenon (Fig. 2, Fig. 3) and in tellurium (Fig. 4), the association of isotopically anomalous high-Z elements with isotopically normal low-Z elements, and the absence of helium and neon in the noble gas component with normal, planetary xenon (Fig. 5, ref. 59) are consistent with the suggestion that our solar system formed directly from the debris of a single supernova[17-19].

We are deeply indebted to Dr. B. Srinivasan for sample preparation and assistance with noble gas analysis, to Dr. David McKown of the University of Missouri Reactor Facility at Columbia and Dr. Phillip Hopke of the University of Illinois at Urbana for use of their counters, and to Dr. Elbert King of the University of Houston for the sample of Allende. This research was supported by the National Science Foundation, NSF-EAR78-09251.

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Table 1 Ratios of stable xenon isotopes

Isotope	Carbon (C-2)				Spinels and Carbon (B-1)			Spinels*	AVCC	Air
	800°	1200°	1600°	Total	800°	1600°	Total	Total		
^{124}Xe	0.497 ± 0.005	0.564 ± 0.008	0.572 ± 0.007	0.548 ± 0.004	0.476 ± 0.006	0.485 ± 0.008	0.478 ± 0.006	0.355 ± 0.018	0.459	0.357
^{126}Xe	0.447 ± 0.008	0.458 ± 0.007	0.456 ± 0.006	0.454 ± 0.005	0.417 ± 0.005	0.409 ± 0.018	0.415 ± 0.008	0.348 ± 0.024	0.410	0.335
^{128}Xe	8.29 ± 0.08	8.42 ± 0.08	8.44 ± 0.11	8.39 ± 0.06	8.29 ± 0.06	8.02 ± 0.17	8.24 ± 0.08	7.97 ± 0.24	8.20	7.14
^{129}Xe	140.3 ± 0.8	106.1 ± 0.6	105.2 ± 0.6	115.2 ± 0.4	116.6 ± 0.5	102.3 ± 0.6	113.8 ± 0.5	111.4 ± 1.6	≈ 100	98.3
^{130}Xe	16.13 ± 0.11	15.90 ± 0.09	15.98 ± 0.09	15.99 ± 0.07	16.28 ± 0.09	15.84 ± 0.10	16.19 ± 0.09	16.55 ± 0.28	16.08	15.17
^{131}Xe	81.55 ± 0.26	82.24 ± 0.29	82.21 ± 0.37	82.04 ± 0.23	81.95 ± 0.32	80.86 ± 0.62	81.74 ± 0.38	81.21 ± 1.12	81.7	78.77
^{132}Xe	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100
^{134}Xe	38.44 ± 0.15	45.21 ± 0.24	43.92 ± 0.17	42.98 ± 0.12	38.15 ± 0.18	40.87 ± 0.18	38.68 ± 0.18	31.16 ± 0.54	38.2	38.82
^{136}Xe	32.41 ± 0.23	42.74 ± 0.30	40.97 ± 0.20	39.39 ± 0.12	32.07 ± 0.22	36.13 ± 0.29	32.86 ± 0.23	21.45 ± 0.67	32.1	32.99
^{132}Xe content (cc STP/g) x 10^{-8}	1.22	1.94	1.29	4.45	2.60	0.62	3.22	2.17	----	-----

*(Amount of isotope in spinels) = (Amount of isotope in B-1) - (Fractional wt. of C in B) $\left(\frac{\text{wt. of B-1}}{\text{wt. of C-2}}\right)$ (Amount of isotope in C-2)

Table 2 Ratios of stable krypton isotopes

Isotope	Carbon (C-2)				Spinels and Carbon (B-1)			Spinels*	AVCC	Air
	800°	1200°	1600°	Total	800°	1600°	Total	Total		
^{80}Kr	4.22 ± 0.06	3.95 ± 0.04	3.92 ± 0.07	4.05 ± 0.04	4.09 ± 0.07	3.87 ± 0.04	4.02 ± 0.06	3.98 ± 0.20	3.92	3.96
^{82}Kr	20.45 ± 0.17	19.72 ± 0.05	19.32 ± 0.03	19.88 ± 0.09	20.25 ± 0.28	19.75 ± 0.17	20.09 ± 0.25	20.51 ± 0.75	20.14	20.21
^{83}Kr	20.42 ± 0.15	20.11 ± 0.11	20.00 ± 0.10	20.20 ± 0.12	20.17 ± 0.10	19.92 ± 0.08	20.09 ± 0.09	19.88 ± 0.36	20.17	20.16
^{84}Kr	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100	≡ 100
^{86}Kr	30.33 ± 0.27	31.44 ± 0.15	31.48 ± 0.19	30.91 ± 0.21	30.55 ± 0.14	31.24 ± 0.15	30.77 ± 0.14	30.49 ± 0.59	30.98	30.55
^{84}Kr content (cc STP/g) x 10^{-8}	4.13	2.89	3.43	10.45	4.98	2.28	7.26	4.53	-----	-----

*See footnote for Table 1 on method used to calculate amount of each isotope in spinels.

Table 3 Ratios of γ-ray intensities detected from activated tellurium isotopes

Isotopes	Carbon: Terr.	Spinels: Terr.	Carbon: Spinels	γ-Energies Detected	Half-life Observed
^{121}Te	106.9 ± 2.6	102.3 ± 1.9	105.4 ± 1.4	0.573 Mev	20-30 d.
^{123}Te	104.3 ± 2.5	100.1 ± 2.4	103.7 ± 2.2	0.159 Mev	120 d.
^{125}Te	97.6 ± 0.5	102.4 ± 0.6	95.4 ± 0.5	0.035 Mev*	58 d.
^{127}Te	105.1 ± 4.1	100.4 ± 3.9	103.4 ± 3.6	0.418 Mev	110 d.
^{129}Te	≡ 100.0	≡ 100.0	≡ 100.0	0.696 Mev, 0.027 Mev*	34 d.
^{131}Te	111.4 ± 4.0	117.5 ± 3.4	94.8 ± 1.7	0.775 Mev, 0.850 Mev	30 hr.

*Counted with intrinsic Ge detector.

Figure 1

Isotopic compositions of xenon in Allende's spinels (□) and carbon (■) normalized to AVCC xenon (○) in the manner given by equation (1). Since ^{129}Xe may contain the *in situ* decay product of ^{129}I, it is not used to define isotopic abundance pattern of trapped xenon. Carbon is enriched in r- and p-products of xenon, spinels are depleted in these, and bulk AVCC xenon has an intermediate abundance pattern for the eight nonradiogenic isotopes of xenon.

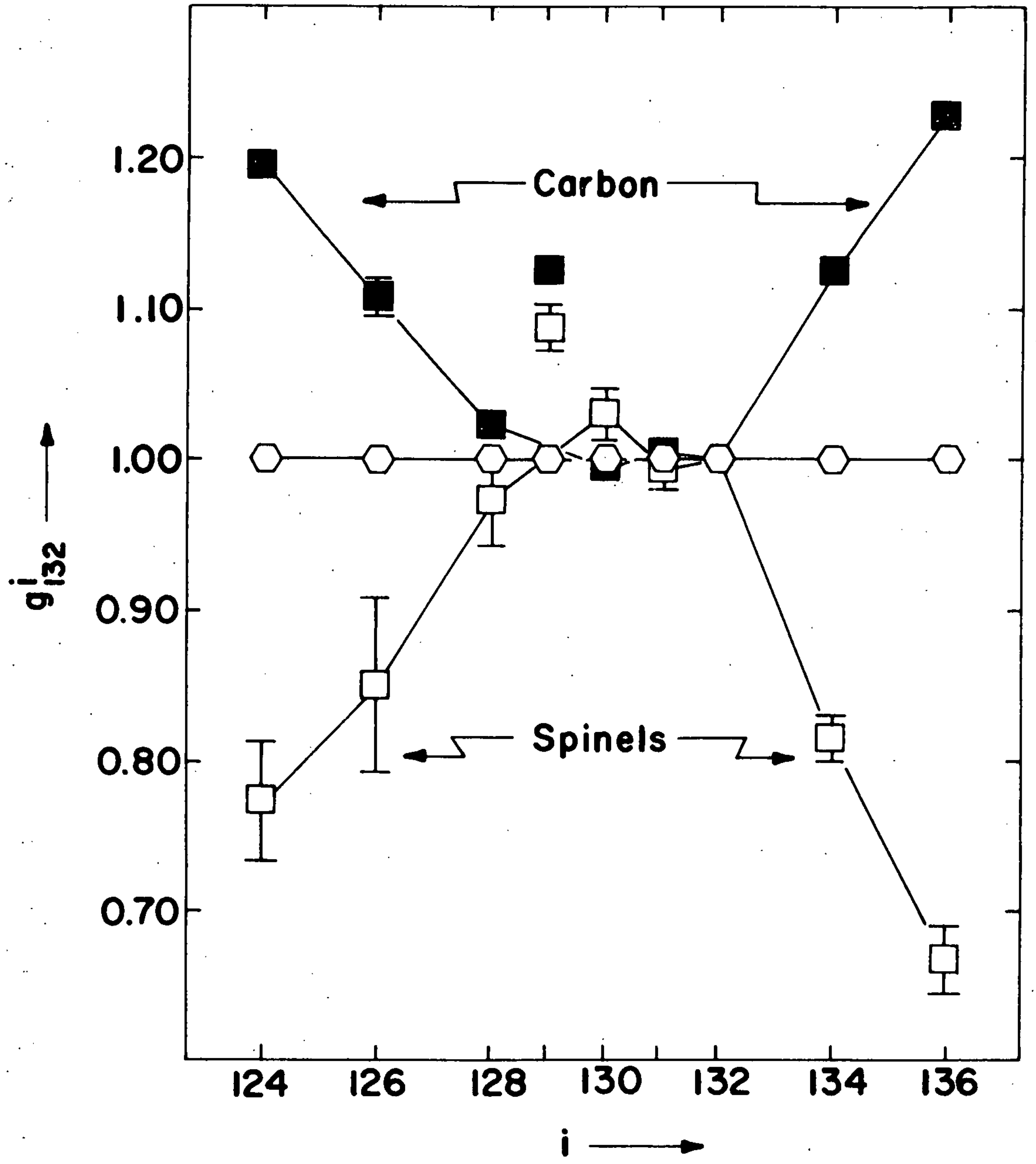
1.20
1.10
1.00
0.90
0.80
0.70
g^i_{132}
Carbon
Spinels
124
126
128
130
132
134
136
i

Figure 1

Figure 2

Variations in abundances of the lightest and heaviest isotopes of xenon released by stepwise heating of Allende's residues. Xenon isotopes released from the mixed residue of carbon and spinels (◪) lie along line *a*, xenon isotopes from the carbon residue (■) lie along line *b*, and xenon isotopes in other residues of Allende are reported[26,27] to lie along the dashed line connecting "fission-free" or "trapped" xenon (○) and "strange" xenon generated by fission and mass fractionation.

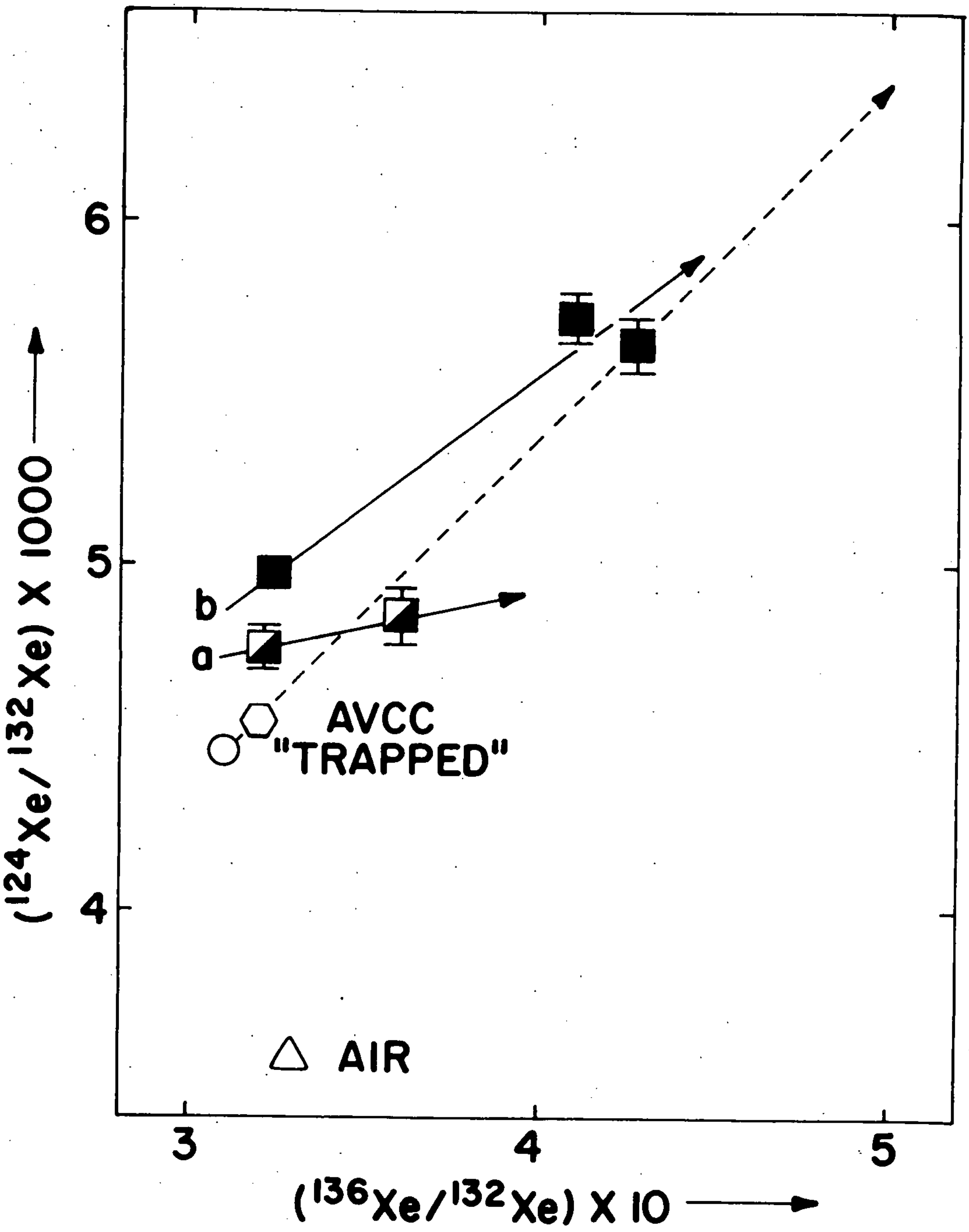
6
5
4
3
4
5
b
a
AVCC
"TRAPPED"
AIR
$(^{124}Xe/^{132}Xe) \times 1000$
$(^{136}Xe/^{132}Xe) \times 10$

Figure 2

Figure 3

Ratios among three xenon isotopes produced by the r-, p-, and s-processes of nucleosynthesis. a, A comparison of trends expected by the addition to AVCC xenon of pure nucleogenetic components from the r-, p-, and s-processes (dark arrows) with trends reported from earlier analyses[26,27] on Allende residues (dashed line); b, trends observed in this study for xenon released by stepwise heating of Allende's carbon (C-2) and carbon plus spinels (B-1); c, trends observed in this study for total xenon in Allende residues; d, the trend observed in an earlier study[26] on xenon released by stepwise heating of one Allende residue (●); e, trends observed in another study[27] on xenon released by stepwise heating of three Allende residues (○,●); f, trends observed[15] in xenon released by stepwise heating of a Murchison residue (●). Other symbols are defined in Figs. 1 and 2. All error limits greater than the size of the symbol are shown, except for the two temperature fractions of Murchison (Fig. 4f) with low values of ^{124}Xe/^{130}Xe. Error limits were not reported[15] on the latter.

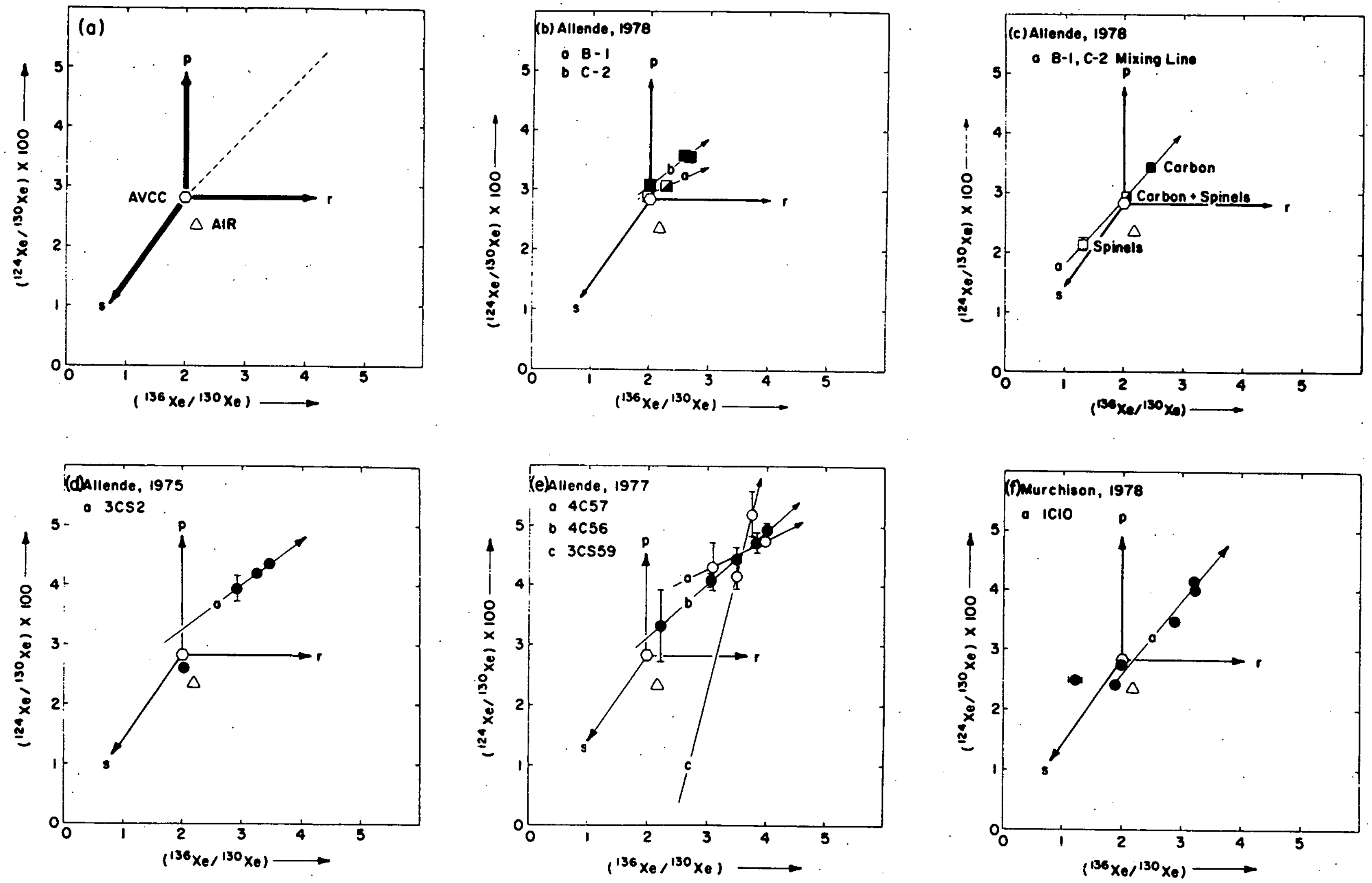
(a)
AVCC
AIR
p
r
s
(b) Allende, 1978
a B-1
b C-2
(c) Allende, 1978
a B-1, C-2 Mixing Line
Carbon
Carbon + Spinels
Spinels
(d) Allende, 1975
a 3CS2
(e) Allende, 1977
a 4C57
b 4C56
c 3CS59
(f) Murchison, 1978
a 1C10
$(^{124}Xe/^{130}Xe) \times 100$
$(^{136}Xe/^{130}Xe)$
0
1
2
3
4
5

Figure 3

Figure 4

Isotopic compositions of tellurium in Allende's carbon (■) and spinels (□) normalized to that in the tellurium monitor (△) in the manner indicated by equation (2). Since ^{126}Te may contain the in situ decay product of ^{126}Sn, it is not used to define the isotopic abundance pattern of trapped tellurium. a, The anomaly pattern of tellurium isotopes in carbon resembles that observed in xenon isotopes (Fig. 1) in carbon; b, the anomaly pattern of tellurium isotopes in spinels reveals an enrichment of the s-product, ^{124}Te, and the r-product, ^{130}Te.

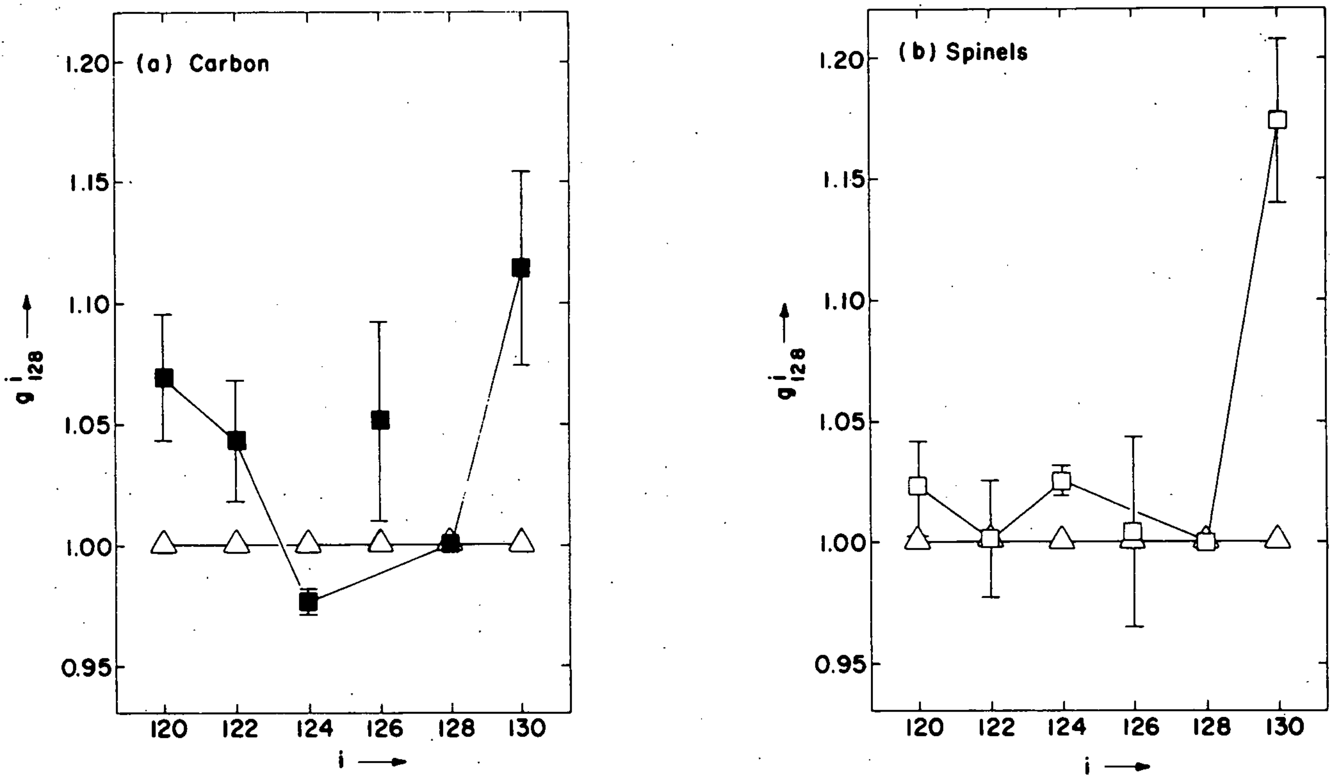
(a) Carbon
(b) Spinels
1.20
1.15
1.10
1.05
1.00
0.95
g^i_{128} →
120
122
124
126
128
130
i →

Figure 4

Figure 5

Correlations between values of the isotopic ratio, $^{136}Xe/^{132}Xe$, and elemental ratios of (low-Z/high-Z) noble gases in residues of Allende[26]. Measured values of elemental ratios are multiplied by constants so that all six ratios can be shown together. Values of ($^{20}Ne/^{132}Xe$) x 60 are depicted by (■), ($^{4}He/^{132}Xe$) x 0.25 by (△), ($^{20}Ne/^{36}Ar$) x 4,000 by (●), ($^{20}Ne/^{84}Kr$) x 25 by (□), ($^{4}He/^{36}Ar$) x 10 by (▲), and ($^{4}He/^{84}Kr$) x 0.04 by (○). The correlations yield zero values for the elemental abundances of helium and neon at a value of $^{136}Xe/^{132}Xe = 0.32 \pm 0.01$. This corresponds to the $^{136}Xe/^{132}Xe$ ratio of bulk xenon in air, carbonaceous chondrites, and ureilites, as shown at $^{136}Xe/^{132}Xe = 0.330$, 0.322 and 0.313, respectively.

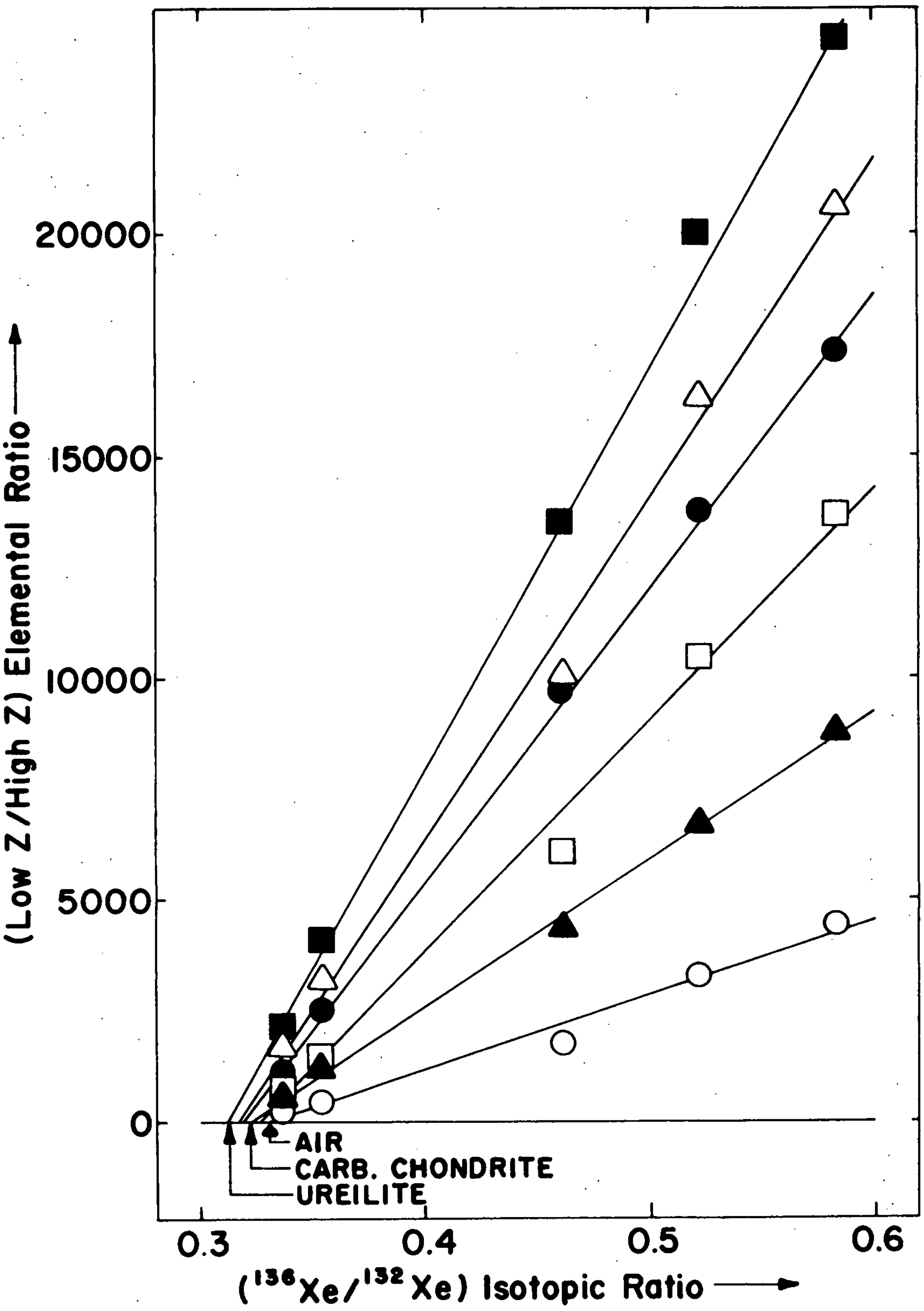
(Low Z/High Z) Elemental Ratio
0
5000
10000
15000
20000
0.3
0.4
0.5
0.6
(^{136}Xe/^{132}Xe) Isotopic Ratio
AIR
CARB. CHONDRITE
UREILITE

Figure 5

VITA

Robert VanGorder Ballad was born on July 26, 1945 in Pittsburgh, Pennsylvania. He received his primary education in Houston, Texas and his secondary education in Park Forest, Illinois. He entered Monmouth College, Illinois, in 1964 and graduated with a B.A. degree in chemistry in 1968. He then entered graduate school and received a M.S. degree from Wayne State University, Detroit, Michigan in December 1970. He entered the University of Missouri-Rolla and worked for two years on ion flotation. He left school for about a year, spent partly in traveling, and then returned to the University of Missouri-Rolla in January 1974 for the work presented in this dissertation.