

THE EARLY SOLAR SYSTEM ABUNDANCE OF  
 $^{244}\text{Pu}$  AS INFERRED FROM THE ST. SEVERIN CHONDRITE

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#### ABSTRACT \*

We describe the analysis of Xe released in stepwise heating of neutron-irradiated samples of the St. Severin chondrite. This analysis indicates that at the time of formation of most chondritic meteorites, approximately  $4.56 \times 10^9$  years ago, the atomic ratio of  $^{244}\text{Pu}/^{238}\text{U}$  was  $0.0068 \pm 0.0010$  in chondritic meteorites. We believe that this value is more reliable than that inferred from earlier analyses of St. Severin. We feel that this value is currently the best available estimate for the early solar system abundance of  $^{244}\text{Pu}$ .

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## INTRODUCTION

$^{244}\text{Pu}$  is a short-lived (half-life 82 Ma) radionuclide which was present in significant quantities early in the history of the solar system (cf. Hohenberg et al., 1967; Alexander et al., 1971; Drozd et al., 1977). Our concern in this paper is in establishing a representative early solar system abundance for  $^{244}\text{Pu}$ .

It is desirable to characterize the abundance of a given isotope by its abundance relative to another isotope of the same element in order to avoid problems due to chemical fractionation. For  $^{244}\text{Pu}$  this is not possible, since no other isotope of Pu has a half-life long enough to have survived the interval between nucleosynthesis and incorporation into solids in the solar system. Normalization to an isotope of some other element is required, and the customary choice is  $^{238}\text{U}$ . One reason for the use of  $^{238}\text{U}$  is convenience, since U abundance is measurable by a variety of techniques. A more fundamental reason is that  $^{244}\text{Pu}$  and  $^{238}\text{U}$  not only are produced in the same nucleosynthesis process, but are also close in mass, so that their relative nucleosynthetic production ratio can be predicted with greater confidence.

Two different experimental techniques have been used to measure the  $^{244}\text{Pu}/^{238}\text{U}$  ratio through fission Xe accumulation. One, which we will term the direct method, is the straightforward approach of measuring the absolute concentration of fission Xe. This is then combined with a corresponding absolute U concentration. The second, which we will term the irradiation method, is based on neutron irradiation of the specimen to

induce fissions of  $^{235}\text{U}$ . Since the composition of  $^{235}\text{U}$  fission Xe is different from that of  $^{244}\text{Pu}$  fission Xe, the composition of the composite fission Xe is a function of the  $^{244}\text{Pu}/\text{U}$  ratio. The measurement of the composite fission Xe composition thus yields the  $^{244}\text{Pu}/^{238}\text{U}$  ratio. The composite fission Xe composition can be calculated from the total sample composition, in which case the composition of non-fission Xe must be assumed or otherwise determined. It can also be based on stepwise-heating isotopic correlation analysis, which does not require assumption of the non-fission Xe composition but does require the chemical coherence of Pu and U. Specifically, that equal fractions of  $^{244}\text{Pu}$ -derived and  $^{238}\text{U}$ -derived fission Xe are released in each of the temperature steps used in the correlation analysis.

In practice, the principal limitation on the precision and reliability of experimental analysis by either method is often the difficulty of resolving fission Xe from other Xe components present in the same specimen: primordial trapped Xe and cosmic ray produced Xe (spallation Xe). All  $^{244}\text{Pu}$  measurements have a systematic uncertainty because of uncertainty in the fission branching ratio, and all those based on Xe are further uncertain by the uncertainty in the Xe yields in fission. These two sources of uncertainty will not affect the comparison of abundance measurements in different samples but they do contribute to uncertainty in the absolute value of  $^{244}\text{Pu}/^{238}\text{U}$ .

It is important to identify two classes of samples on which  $^{244}\text{Pu}$  measurements have been performed: whole rock undifferentiated (chondritic) meteorites, and all others. Whole rock analyses of chondrites are

important in determining the solar system abundance of  $^{244}\text{Pu}$  because it is generally acknowledged that these meteorites are the best representatives of solar system composition for nonvolatile elements. Whole rock chondrite analyses of  $^{244}\text{Pu}/^{238}\text{U}$  are experimentally difficult because of a low abundance of  $^{244}\text{Pu}$  fission Xe to other Xe components. To date only one usefully precise  $^{244}\text{Pu}/^{238}\text{U}$  ratio has been reported for a whole rock chondrite: Podosek (1970a) reported a  $^{244}\text{Pu}/^{238}\text{U}$  ratio for St. Severin based on the isotope-correlation method. Podosek (1972) reported a revised value, based on different parameters but the same experimental analysis, of  $^{244}\text{Pu}/^{238}\text{U} = 0.0154 \pm 0.0014$ ; this figure is often cited as the best available estimate for the solar system abundance of  $^{244}\text{Pu}$ .

The "other" category of measurements are for samples which are, by definition, not chondritic, and thus are chemically unrepresentative of solar system abundance in one way or another. Examples include specific mineral phases within ordinary chondrites, refractory element rich inclusions and igneous differentiate meteorites. These "other" samples are frequently poor in volatiles, including trapped Xe, and/or rich in refractories, including Pu and U, with the result that measurement of  $^{244}\text{Pu}$  abundance and/or  $^{244}\text{Pu}/^{238}\text{U}$  is easier and more reliable than in the case of whole rock chondrites. However these "other" samples have undergone significant chemical differentiation and their relevance of such measurements to the solar system abundance of  $^{244}\text{Pu}$  is less certain.

It can be argued (e.g. Ganapathy and Grossman, 1976 ; Lugmair and Marti, 1977 ; Marti et al., 1977 ; Jones, 1982) that analyses of appropriate "other" samples can be used to determine the solar system abundance of  $^{244}\text{Pu}$ . The basis of such argument is that in the appropriate sample, Pu has not been chemically fractionated from some other element, either U itself or another, notably Nd, which can be used as an index of solar system abundance.

While these "other" measurements are more model dependent than whole rock chondrite measurements it is clear that they are not consistent with  $^{244}\text{Pu}/^{238}\text{U} = 0.015$ . Lugmair and Marti (1977) and Marti et al. (1977) have argued for a value of  $^{244}\text{Pu}/^{238}\text{U} = 0.004$  based primarily on data from Angra dos Reis where  $^{244}\text{Pu}/^{238}\text{U}$  is obtained by measuring  $^{244}\text{Pu}$  and Nd and multiplying by Nd/U as measured in chondrites.

This controversy has been discussed at length by Jones and Burnett (1979), Burnett et al. (1982) and Jones (1982) with varying conclusions regarding the best estimate of cosmic  $^{244}\text{Pu}/^{238}\text{U}$ . The first two papers prefer  $^{244}\text{Pu}/^{238}\text{U} = 0.015$  and the last paper favors  $^{244}\text{Pu}/^{238}\text{U} = 0.005$ , approximately the value recommended by Lugmair and Marti (1977) and Marti et al. (1977). Because of this discrepancy we have reanalyzed several St. Severin samples. The persistent use of St. Severin can be questioned, since comparable measurements in other chondrites would be of value. The selection of St. Severin is partly a matter of choice and partly a matter of necessity. St. Severin is a good sample for the present investigation because it is a well characterized in terms of petrography and trace elements, and has been extensively studied in terms of

radiogenic isotope systematics, so that a good chronological framework can be established. St. Severin is also suitable in terms of the feasibility of the experiment: it has a modest cosmic ray exposure age and a low concentration of trapped Xe.

#### SAMPLES

St. Severin (LL6 chondrite) has two distinct lithologies (Orcel et al., 1967 ; Wasserburg et al., 1969 ; Jones and Burnett, 1979 ; Hohenberg et al., 1981a) characterized as "light" and "dark". This structure is probably the result of impact brecciation early in St. Severin's history (Ashworth and Barber, 1977). The dark lithology consists of large dark angular fragments ( $>1 \text{ cm}^3$ ) imbedded in a light colored "matrix" of crushed material which constitutes the light lithology. Separations of light and dark material were made by slicing a large piece (20 g from fragment D, cf. Orcel et al., 1967) into 1 mm thick slabs. In these slabs it was easy to identify and separate the two lithologies. Additional samples were gently crushed by hand under methanol with stainless steel tools. First, troilite grains were hand picked and then a sample enriched in whitlockite was obtained with a Franz magnetic separator (cf. Lewis, 1975). The remaining material was treated with nitric acid in an attempt to remove phosphates. The remaining coarser material was magnetically separated to produce a plagioclase rich and plagioclase poor sample. The samples analyzed are summarized in Table 1.

## EXPERIMENTAL PROCEDURES AND RESULTS

Chips of meteorite samples were sealed under vacuum in quartz vials and loaded into an aluminum irradiation capsule. The capsule was irradiated in 1979 in the rotating can facility in the reflector of the Research Reactor Facility at the University of Missouri (Columbia). The laboratory designation for this irradiation is SLC5.

Spatial variation in neutron fluence was determined by measuring induced activities in Co-doped Al wires included in each vial. Measurements indicate negligible fluence variations ( $< 1\%$ ). These measurements, along with post irradiation sample handling procedures are discussed by Kennedy (1981).

Noble gases were released from the samples by stepwise heating (RF induction) for one hour at each step (Two hours for the unirradiated samples) in a Mo crucible. Temperatures were calibrated with an optical pyrometer and are probably accurate to  $50^{\circ}\text{C}$ . The noble gases were purified by successive exposure to freshly deposited Ti films.

Ar and Xe were separated by selective adsorption on activated charcoal. Two mass spectrometers connected by a common line were used for gas analysis: MS2 (Drozd, 1974) was used for Ar and MS3 (Hohenberg, 1980) was used for Xe. Additional details of the operating procedures are given by Hudson (1981) and Kennedy (1981).

A full tabulation of the raw data is given by Hudson (1981) and will not be repeated here.

## DATA REDUCTION AND ANALYSIS

For both samples and neutron irradiation monitors, a number of corrections must be applied to the data in order to identify the reaction products of interest. These include corrections for procedural blanks, cosmic ray spallation, natural fission and a variety of reactions induced in the neutron irradiation. In general, the application of these corrections does not introduce any significant uncertainty into the evaluations. Either the corrections are straightforward or the effects are small enough that the uncertainty in the correction is not important. Details of the computations and algorithms are discussed by Hudson (1981). All calculations recognize that uncertainties may be partially correlated. Figure 1 summarizes the decomposition of the observed compositions into their constituent parts.

Irradiation interference corrections

The neutron irradiation produces  $^{130}\text{Xe}$  and  $^{132}\text{Xe}$  from neutron capture on Xe. Corrections are made using thermal neutron cross-sections of 20 and 90 barns respectively, and the estimated neutron fluence of  $2 \times 10^{19}$  n/cm<sup>2</sup> for SLC5. These corrections are minor (< 1%). Another interference arises from (n,2n) reactions on  $^{127}\text{I}$ . It is possible, using the data of another SLC5 sample (St. Sauveur), to deduce the ratio of  $^{126}\text{Xe}/^{128}\text{Xe}$  from neutron reactions on I. Kennedy (1981) discusses this problem and concludes  $(^{126}\text{Xe}/^{128}\text{Xe})_{\text{I}} = (2.9 \pm 0.1) \times$

$10^{-5}$ . This value can be used to correct the observed  $^{126}\text{Xe}$  based on I-derived  $^{128}\text{Xe}$ . These corrections range up to 10% for extractions below  $1000^{\circ}\text{C}$  but are less than 1% for higher temperatures.

### Spallation Xe

The composition of Ba and REE (Rare Earth Element) spallation Xe in the Angra dos Reis achondrite are well determined and provide a model for meteoritic spallation Xe (Hohenberg et al., 1981b). While the composition of Ba and REE spallation Xe are expected to vary to some extent from sample to sample due to shielding differences, general agreement is seen between the composition of St. Severin spallation Xe and that predicted by the Angra dos Reis Xe spallation systematics. In particular, the total sample values of  $^{124}\text{Xe}/^{126}\text{Xe}$  in spallation are in agreement with the chondritic value of 0.60 predicted by the Angra dos Reis data.

Figures 2 and 3 show the mixing of trapped Xe and spallation Xe in the St. Severin samples. The value of  $^{124}\text{Xe}/^{126}\text{Xe}$  in the composite spallation component is determined by the intersection of the Ba-REE spallation mixing line and the line connecting the trapped composition and the observed composition. For gases released below  $1100^{\circ}\text{C}$ , air has been used as the trapped composition. This procedure is ad hoc but it leads to a consistent decomposition and the uncertainties associated with it are small since the low temperature release constitutes a small portion of the total Xe.

Two points should be made regarding the inferred spallation compositions: considerable variation is seen, ranging up to the extremes allowed by the AVCC-Ba-REE endmembers, and agreement is seen between the total compositions of the irradiated and unirradiated samples, although their temperature release patterns are not identical.

The value of  $^{124}\text{Xe}/^{126}\text{Xe}$  in spallation at each temperature determines the ratio of Ba to REE contributions and thus the composition of spallation Xe at all isotopes, given the Angra dos Reis model of Hohenberg et al. (1981b). With this, spallation contributions can be subtracted at all isotopes.

#### Fission from U

The production of Xe from neutron induced fission of  $^{235}\text{U}$  was calibrated using a sample of NBS 0.02 ppm series trace element glass (72 ppb U). Xenon data for this sample is given in Kennedy (1981). The U conversion efficiency for SLC5 is  $(3.8 \pm 0.4) \times 10^{-4} \text{ cm}^3 \text{STP}^{136}\text{Xe/gU}$ . For proper comparison to meteorite samples, the value has had added, (trivial) contributions from  $^{238}\text{U}$  as if the sample had a 4.55 Ga decay period.

It is instructive to compare this value to the conversion efficiency computed on the basis of  $^{127}\text{I}$  conversion assuming a thermal neutron spectrum. A whole rock sample of Bjurbole irradiated in SLC5 gave  $^{128}\text{Xe}/^{129}\text{Xe}$  (from iodine) =  $0.9741 \pm 0.0030$ . Assuming this corresponds to  $^{129}\text{I}/^{127}\text{I}$  of  $1.095 \times 10^{-4}$  (Hohenberg and Kennedy, 1981), the implied thermal fluence is  $2.0 \times 10^{19} \text{ n/cm}^2$ . This fluence, along with

a fission cross-section of 584 barns and a 7.8% yield at  $^{134}\text{Xe}$  implies  $8.8 \times 10^{-4} \text{ cm}^3\text{STP}^{136}\text{Xe}/\text{gU}$ , roughly a factor of two greater than that directly measured. This presumably reflects that about half of the  $^{127}\text{I}$  neutron captures are due to epithermal neutrons.

#### Fission from Pu

After the corrections previously described,  $^{130}\text{Xe}$ ,  $^{132}\text{Xe}$ ,  $^{134}\text{Xe}$  and  $^{136}\text{Xe}$  are mixtures of trapped Xe and fission Xe. In the unirradiated samples, fission Xe is primarily from spontaneous fission of  $^{244}\text{Pu}$  with small contributions from  $^{238}\text{U}$  spontaneous fission. Corrections for  $^{238}\text{U}$  fission are based on estimated Pu/U ratios for each extraction; for  $^{244}\text{Pu}/^{238}\text{U} = 0.005$ , 3.7% of the total fission  $^{136}\text{Xe}$  is due to  $^{238}\text{U}$ . A 50% error has been assumed in this correction.

Figures 4 and 5 show the mixing of trapped Xe and  $^{244}\text{Pu}$  fission Xe for this work and the whitlockite data of Lewis (1975). The intercepts of the mixing lines give the composition of  $^{244}\text{Pu}$  fission since  $^{130}\text{Xe}$  is not significantly produced in fission. Only data from extractions at  $1100^\circ\text{C}$  and above have been used in order to avoid atmospheric Xe contamination. Also included are the whitlockite data of Lewis (1975), corrected as described above. The addition of these data does not change the inferred  $^{244}\text{Pu}$  composition but reduces the calculated uncertainties by about 20%. Table 3 gives the computed  $^{244}\text{Pu}$  fission Xe composition and compares it to previous determinations.

### Trapped Xe

To resolve Pu and U fission Xe contributions in a single Xe composition, we must know the composition of the trapped Xe component. Compositions such as AVCC (Eugster et al., 1967) represent, perhaps, a reasonable a priori choice for the trapped composition. As seen in figs. 4 and 5 however, data for Xe extracted at high temperatures from the unirradiated, nitric acid etched samples are inconsistent with AVCC as a trapped component. In general, the trapped Xe composition must lie on the trapped-fission mixing lines shown in figs 4 and 5 and must be to the right of the most fission-free point (1650°C extraction of SM, 25% of the Xe in sample SM).

A similar situation exists for the irradiated samples, but in this case Pu/U coherence is required in order to produce a single fission composition and a well defined trapped-fission mixing line. Such a Pu/U coherence is apparently observed in the high ( $\geq 1175^{\circ}\text{C}$ ) temperature extractions of the dark lithology (also, but to a lesser extent in the light lithology). Figure 6 and 7 shows a least squares line fit to the high temperature data of sample SD5. This mixing line must have the same trapped component as that for the unirradiated samples and thus the intersection of these lines determines the trapped composition. This line is shown in comparison to the data of sample SL5 in figure 7.

The intersection value of  $^{130}\text{Xe}/^{136}\text{Xe}$  matches that observed in the 1650°C extraction of SM, the most "fission-free" point. This value combined with the unirradiated fission-trapped mixing lines then determines the trapped composition at  $^{130}\text{-}^{136}\text{Xe}$  and is given in Table 3.

Our immediate interest in the trapped Xe composition is as necessary parameter in the evaluation of the  $^{244}\text{Pu}/^{238}\text{U}$  ratio in a single release fraction. A detailed discussion of the trapped Xe composition and its possible cosmochemical significance is beyond the scope of this paper. It should be noted, however, that in view of the complexities involved in the isotopic structure of Xe in various samples of the solar system (cf. Podosek, 1978; Pepin and Phinney, 1982) no presumptive case can be made that the composition of trapped Xe in St. Severin must or even should match that of other kinds of meteorites. Given this premise, trapped Xe isotopic structures for ordinary chondrites must be regarded as largely unexplored territory.

#### Pu/U Ratios

Given data corrected for spallation and irradiation interferences, and the compositions of U fission Xe,  $^{244}\text{Pu}$  fission Xe, and trapped Xe (Table 3, air Xe for temperatures  $\leq 1000^\circ\text{C}$ ), each temperature fraction can be resolved into the two fission and one trapped components. The results of this calculation for both SL5 and SD5 are shown in Figure 8.

Large variations of  $^{244}\text{Pu}/^{238}\text{U}$  are evident in Figure 8. (also see Figures 6 and 7). The basic qualitative feature is higher  $^{244}\text{Pu}/^{238}\text{U}$  at intermediate ( $1000^\circ\text{C} - 1200^\circ\text{C}$ ) temperatures. The variations in  $^{244}\text{Pu}/^{238}\text{U}$  are evidently manifestations of chemical fractionation of Pu relative to U on a microscopic scale. The obvious (but unverified) assumption is that the lower temperature peak in  $^{244}\text{Pu}/^{238}\text{U}$  reflect gas release from whitlockite. Since even the highest  $^{244}\text{Pu}/^{238}\text{U}$  in

Fig. 8 is a factor of two-to-several below values obtained on separated whitlockite from St. Severin, it seems unlikely that the low temperature peak reflects solely a whitlockite contribution.

In view of the significant variations in  $^{244}\text{Pu}/^{238}\text{U}$  in stepwise heating, it is clearly inappropriate to estimate it on the basis of a least-squares isotopic correlation fit. We therefore compute bulk sample ratios by integration of the stepwise heating resolution. These values for initial  $^{244}\text{Pu}/^{238}\text{U}$  are  $0.0062 \pm 0.0008$  for the light lithology (SL5) and  $0.0051 \pm 0.0005$  for the dark lithology (SD5). The quoted errors are based on uncertainties in the raw data, the spallation corrections, the endmember compositions; they do not include uncertainties in the decay parameters of  $^{244}\text{Pu}$  or in the irradiation calibration for  $^{235}\text{U}$  fission.

#### Epoch

Because the lifetime of  $^{244}\text{Pu}$  is relatively short, it is necessary to consider the epoch to which the measurement described above refer. A significant marker epoch in the early solar system, in the sense of an early event whose age can be determined absolutely, is the chemical fractionation of U from Pb in coarse-grained inclusions in Allende, dated by  $^{207}\text{Pb}$ - $^{206}\text{Pb}$  ages of  $4.56 \pm 0.01$  Ga (e.g. Chen and Tilton, 1976; Chen and Wasserburg, 1981). Unfortunately, it is rather difficult to relate noble gas data to this or similar ages. Therefore (following Podosek, 1970b) we will relate gas data, to the extent possible, to Xe isotopic closure in the chondrite Bjurböle, specifically as defined by the

$^{129}\text{I}$ - $^{129}\text{Xe}$  method. It is difficult to assign a precise and unambiguous absolute age to this event, but presumably it is not much different than 4.56 Ga.

There is no single age which can be assigned for the "formation" of St. Severin: different chronometers have different definitions of "formation" or "closure." Its  $^{207}\text{Pb}$ - $^{206}\text{Pb}$  age is indistinguishable (within 10 Ma) from that of the Allende inclusions cited above (Mahnes et al., 1978; Chen and Wasserburg, 1981). Both elevated initial  $^{87}\text{Sr}/^{87}\text{Sr}$  (Mahnes et al., 1978) and absolute  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  ages (e.g. Hohenberg et al., 1981a), however, indicate "formation" at considerably later times, more than 100 Ma later. This spread of ages presumably reflects the different responses of the different chronometers to metamorphism in the parent body of St. Severin, which spanned at least  $10^8$  years. Uncertainty in the epoch to which the  $^{244}\text{Pu}$  abundances refer might thus be comparably to the half life of  $^{244}\text{Pu}$ .

Presumably, the independent chronometer most likely to date the time of retention of fission Xe is the  $^{129}\text{I}$ - $^{129}\text{Xe}$  age. It might be argued, however, that even though the daughter element is the same in both cases, closure for radiogenic  $^{129}\text{Xe}$  need not occur at the same time as closure for fission  $^{136}\text{Xe}$  if their parent distributions are significantly different. It is difficult to refute such an argument, but equally difficult to defend any alternative chronology. That (high temperature) radiogenic  $^{129}\text{Xe}$  and fission Xe should date the same event is suggested by the similarity in the high-temperature ( $>1000^\circ\text{C}$ ) release patterns of Xe products from irradiation of I and U (Fig. 1). At any rate, we will

assume that the time at which the samples began to retain  $^{244}\text{Pu}$  fission Xe is best dated by the  $^{129}\text{I}$ - $^{129}\text{Xe}$  age obtained in the same experiment.

It can be argued that the metamorphism occurred as a closed system, and that radiogenic decay products would accumulate in spite of microscopic mobility. That this is the case, at least to some extent, is indicated by the elevation of trapped  $^{129}\text{Xe}/^{130}\text{Xe}$  (equivalent to the elevation of initial  $^{87}\text{Sr}/^{86}\text{Sr}$ ) in these samples. In such case, however, fission Xe, like radiogenic  $^{129}\text{Xe}$ , would be homogenized with any ambient (now trapped) Xe. Since our trapped Xe composition is inferred from observed isotopic structures rather than assumed on the basis of any analogy, accumulation of fission Xe in closed system metamorphism would mean that primordial Xe composition is even further back along the fission mixing line (Fig. 4 and 5). Our algorithm resolves in situ fission Xe, and the corresponding  $^{244}\text{Pu}$  abundance is that at the time of microscopic Xe closure, not retention of Xe in the bulk sample.

The  $^{129}\text{I}$ - $^{129}\text{Xe}$  ages of light (SL5) and dark (SD5) lithologies are  $9 \pm 1$  and  $36 \pm 2$  Ma after Bjurbole (Hudson, 1981), respectively. Correcting the corresponding  $^{244}\text{Pu}/^{238}\text{U}$  ratios for decay during these intervals yields initial (at the time of radiogenic  $^{129}\text{Xe}$  closure in Bjurbole)  $^{244}\text{Pu}/^{238}\text{U}$  ratios of 0.0067 for the light lithology and 0.0069 for the dark lithology. Although the uncertainties are large, it is interesting to note that the relative I-Xe age agrees with the relative Pu-U-Xe age.

Our best estimate for the "cosmic" value of initial  $^{244}\text{Pu}/^{238}\text{U}$  is thus  $0.0068 \pm 0.0010$ . The stated error includes all uncertainties associated with the experimental measurements, corrections, and resolutions, and also the uncertainty in the irradiation calibration for  $^{235}\text{U}$  fission; it does not include any uncertainties associated with the assumed decay properties of  $^{244}\text{Pu}$  (fission branching ratio and  $^{136}\text{Xe}$  yield) or the final adjustment for epoch.

#### COMPARISONS AND EVALUATIONS

##### St. Severin

In comparison with the Podosek (1970a) analysis, the present measurements are more accurate and the samples better documented, and two independent analyses yield the same result. In the absence of any arguments to the contrary, the present measurements should be given preference on these grounds alone. The discrepancy is more than a factor of two, however, and cannot be attributed solely to improved analytical precision.

One possibility is sampling heterogeneity. Pu and U are highly concentrated in phosphates, and whitlockite in particular evidently enriches Pu relative to U, so analysis of a specimen anomalously enriched in phosphates would lead to an unrepresentative high Pu/U ratio: in short, even bulk chondrite, in gram sized samples, may be liable to trace element fractionation.

A second possibility is failure of the assumed coherence of Pu and U within a given specimen, specifically the assumption that fission Xe from  $^{244}\text{Pu}$  and  $^{235}\text{U}$  will be released in constant proportion in stepwise heating and so define a single fission component whose composition yields the initial  $^{244}\text{Pu}/^{238}\text{U}$  ratio. Studies on the distribution of  $^{244}\text{Pu}$  and U within chondrites indicate that this is a risky assumption. As noted by Jones and Burnett (1979), the 1100°C and 1200°C fractions in the Podosek (1970a) analysis have the greatest enrichment of fission Xe relative to trapped Xe but nevertheless account for only a relatively minor fraction (about 30%) of the total quantity of fission Xe. If the assumption of a single fission component fails, i.e., if the equivalent  $^{244}\text{Pu}/^{238}\text{U}$  ratio is variable in stepwise heating, the greater leverage of the 1000°C and 1200°C points will lead to a weighting of these points in the correlation analysis which is disproportionate in comparison to what they contribute to the total inventory.

A third potential source of discrepancy is incorrect irradiation calibration. Podosek (1970a,b) calculated the extent of  $^{235}\text{U}$  fission in the V2 irradiation from the thermal neutron cross-section and a neutron fluence based on neutron capture by  $^{127}\text{I}$ . Epithermal resonances are more important for  $^{127}\text{I}$  than for  $^{235}\text{U}$ ; if there is a significant epithermal contribution to  $^{127}\text{I}$  this could lead to an overestimate of thermal neutron fluence, thus an overestimate of the extent of  $^{235}\text{U}$  fission, thus an underestimate of U abundance, and thus an overestimate of  $^{244}\text{Pu}/^{238}\text{U}$  ratio. As noted earlier, estimating the extent of  $^{235}\text{U}$  fission in the SLC5 irradiation on the basis of  $^{127}\text{I}$  capture would

produce a discrepancy of more than a factor of two. This is certainly suggestive in terms of the comparison of the Podosek (1970a, 1972) and the present values for  $^{244}\text{Pu}/^{238}\text{U}$ . As noted by Jones and Burnett (1979), it is similarly suggestive that the Podosek (1970a) data corresponds to a U content of about 6 ppb, about a factor of two lower than that determined with more reliable methods. However, it is worth noting that the V2 irradiation, unlike the SLC5 irradiation, was performed far from the reactor core, with considerable shielding which should have effectively thermalized the neutron spectrum.

In summary, we have discussed three effects which might account for the discrepancy between the present and the Podosek (1970a, 1972) values for  $^{244}\text{Pu}/^{238}\text{U}$ . The sense of the effects is such as to support the view that the present estimate is to be preferred to the earlier, but the discussion underscores the difficulty in making reliable measurement of initial  $^{244}\text{Pu}/^{238}\text{U}$ .

Hohenberg et al. (1981a) reported results for St. Severin samples from an earlier irradiation, SLC4. Unfortunately, these data are not very relevant in the present context. Of two samples, one (the light lithology) was analyzed in Al foil in which it was irradiated, and fission Xe from U in the Al foil overwhelmed the fission Xe from the sample. The second (dark lithology) was not analyzed in the irradiated foil; data for this sample are consistent with those from the SLC5 sample, but are not as useful for determination of the  $^{244}\text{Pu}/^{238}\text{U}$  ratio.

Similarly there are additional older analyses of bulk St. Severin samples reported by Wasserburg et al., (1969), Marti et al. (1969) and Alaerts et al. (1979). As with the SLC4 samples these are not especially useful for determining  $^{244}\text{Pu}/^{238}\text{U}$ . However, these data, combined with an assumed trapped Xe composition and an assumed U concentration yield values of  $^{244}\text{Pu}/^{238}\text{U}$  between  $10^{-3}$  and  $10^{-2}$  and thus are more consistent with this work than the 0.015 value of Podosek (1972).

#### Other Measurements

As we have noted, there are not other bulk chondrite analyses of initial  $^{244}\text{Pu}/^{238}\text{U}$  comparable to those for St. Severin. None of the other analyses of neutron-irradiated chondrites by Podosek (1970b), for example, yield irradiation method determinations of useful precision, nor are we aware of any other such analyses which do. Direct method determinations, while less sensitive to analytical uncertainties, have also not yielded results of the necessary precision. Direct method determinations also require assumption of a trapped composition, and in view of uncertainties about trapped Xe composition in ordinary chondrites, as discussed above for the particular case of St. Severin, the need for this assumption introduces an ambiguity whose quantitative importance is difficult to estimate.

Chondrules separated from Bjurbole and Allende (Caffee et al., 1982; Swindle et al., 1983) have been analyzed by the irradiation method. While the data from single chondrules is not sufficiently precise to determine  $^{244}\text{Pu}/^{238}\text{U}$ , the sum of all the gas extracted is precise enough to

yield a meaningful  $^{244}\text{Pu}/^{238}\text{U}$  ratio. Swindle et al., (1983) obtained  $^{244}\text{Pu}/^{238}\text{U} = 0.0073 \pm .0025$  for Bjurbole and  $^{244}\text{Pu}/^{238}\text{U} = 0.0056 \pm .0014$  for Allende. The extent to which Pu and U might be fractionated in these chondrules is uncertain. Chondrules are certainly less chemically fractionated than individual calcium-aluminum inclusions in Allende or separated minerals from St. Severin. Given the greater chemical diversity of spherical objects in Allende as compared to Bjurbole (cf. Swindle et al., 1983), the  $^{244}\text{Pu}/^{238}\text{U}$  value obtained from the Bjurbole chondrules is probably more representative of chondritic abundances.

Phosphates (whitlockite and chlorapatite) have a special relevance in  $^{244}\text{Pu}$  studies. They are typically rather scarce but separable, have low trapped Xe abundances, high abundances of relevant trace elements, and are often sufficiently large to permit track studies. An early analysis (direct method) of whitlockite in St. Severin by Wasserburg et al. (1969), for example, led to an estimate of  $^{244}\text{Pu}/^{238}\text{U} = 0.035$ , which was cited as a candidate cosmic composition. Since the studies of Crozaz (1974) it is evident that phosphates do not provide an unbiased sample of the actinide and REE trace element composition of their host rocks, and in particular that whitlockite enriches Pu relative to U. Kirsten et al. (1978), for example, report direct method analyses for phosphates in 10 chondrites, with  $^{244}\text{Pu}/^{238}\text{U}$  mostly in the range 0.004 - 0.010, but one (Nadiabondi) as high as 0.120, and  $^{244}\text{Pu}/^{238}\text{U}$  typically an order of magnitude higher in whitlockite than in chlorapatite. Lewis (1975) reports an irradiation method analysis of St. Severin whitlockite, obtaining a total  $^{244}\text{Pu}/^{238}\text{U}$  ratio near that of the earlier Wasserburg

et al. (1969) value and explicitly noting a variation in the range 0.017 - 0.045 among gas extractions at different temperatures. It is clear that in general phosphates do not provide an unfractionated Pu/U ratio. St. Severin is probably a case of particularly strong bias, since this meteorite, unlike the case for some other meteorites, the phosphates are almost entirely whitlockite (cf. Jones and Burnett, 1979). Ca-Al-rich inclusions in some carbonaceous chondrites are another class of samples which is readily separable and yields precise  $^{244}\text{Pu}$  abundances. The  $^{244}\text{Pu}/^{238}\text{U}$  ratios in fine-grained inclusions from Allende are very high and it is evident that this reflects a strong chemical fractionation between Pu and U (Podosek and Lewis, 1972). The status of the Ganapathy and Grossman (1976) hypothesis, that average abundances of refractory elements in coarse-grained inclusions should be in cosmic ratio, remains unclear insofar as it applies to Pu and U. Appropriate data are available for only a single inclusion (albeit a large one), for which Drozd et al. (1977) reported  $^{244}\text{Pu}/^{238}\text{U} = 0.016 \pm 0.002$  (direct method); this coincidence with the Podosek (1972) value was taken to support the validity of both that value and the Ganapathy and Grossman hypothesis. In this inclusion most refractory elements, apparently including Pu, are more or less uniformly enriched relative to chondrites, by a factor of about 18 (Drozd et al., 1977; Podosek et al., 1977; also see Jones, 1982). The relevant exception, however, is U, which is also enriched relative to chondrites but not by nearly so great a factor (also see Boynton, 1978). The circumstance that  $^{244}\text{Pu}/^{238}\text{U}$  (direct method) in this inclusion is nearly the same as the Podosek (1972) value for St. Severin must be dismissed as only coincidence.

A number of achondritic meteorites have low trapped Xe concentrations and high refractory trace element concentrations, and so are also particularly suitable for  $^{244}\text{Pu}$  measurements. Fission Xe and/or U concentrations, however, are often variable from one specimen to another by as much as a factor of two. A number of early analyses (see e.g., Sabu and Kuroda, 1967, and also Reynolds, 1968) indicate initial  $^{244}\text{Pu}/^{238}\text{U}$  ratios in the general range of interest here, i.e. about 0.004-0.016, but are not really useful in terms of defining the cosmic ratio within this range. There are in fact rather few cases in which fission Xe, U and light REE data, along with adequate chronological constraints, are available for the same sample.

#### Alternative Normalizations

Interest in elements which might be more coherent (than U) with Pu generally focuses on the light REE. Lugmair and Marti (1977), for example, suggested that Pu is coherent with Nd on the basis of nearly equal Pu/Nd in phosphates and pyroxene in Angra dos Reis, and Marti et al. (1977) have extended these results to Ca-Al-rich inclusions and to Juvinas.

Arguments for coherence of Pu with Nd (or other light REE) can be supported theoretically, in that in normal igneous differentiation processes the relevant distribution coefficients are sufficiently small and similar that chemical fractionation should be minor in appropriate circumstances (cf. Benjamin et al., 1978; Jones, 1982). This is consistent with equality of Pu/Nd in coexisting phases of Angra dos Reis,

but this argument itself cannot be used in support of the thesis that the observed Pu/Nd ratio in Angra dos Reis is the cosmic ratio; the parent body of Angra dos Reis is chemically fractionated relative to cosmic abundances, and in this case the fractionation is evidently nebular (i.e. governed by gas/solid partitioning). It can also be argued, however, that Pu should be relatively coherent with the light REE even in nebular condensation processes (cf. Boynton, 1978), although perhaps with somewhat greater latitude for fractionation.

#### Summary

We conclude that  $^{244}\text{Pu}/^{238}\text{U} = 0.0068 \pm .0010$  is presently the best available estimate of the early solar system abundance of  $^{244}\text{Pu}$ . This value is similar to that recently obtained for a suite of chondrules separated from the Bjurböle chondrite;  $^{244}\text{Pu}/^{238}\text{U} = 0.0073 \pm .0025$  (Swindle et al., 1983).

This value is also similar to the value recommended by Lugmair and Marti (1977) and Marti et al. (1977) of  $^{244}\text{Pu}/^{238}\text{U} = 0.004$  and Jones (1982) of  $^{244}\text{Pu}/^{238}\text{U} = 0.005 \pm .002$ . Whether the value of  $^{244}\text{Pu}/^{238}\text{U} = 0.004$ , obtained principally from Angra dos Reis (Lugmair and Marti, 1977), is significantly different from 0.007 obtained here for St. Severin is difficult to evaluate at this time.

While even gram size samples of bulk chondrites are not guaranteed to have cosmic ratios of refractory elements, they are the best available material for which an a priori assertion of non-fractionation of refractory elements can be made. We thus consider that the estimation of

the cosmic abundance of  $^{244}\text{Pu}$  must be made on bulk chondrite measurements.

#### IMPLICATIONS

The cosmic abundance of  $^{244}\text{Pu}$  is an important parameter in a variety of geochemical and cosmochemical problems. If, as we consider appropriate, the best estimate for the cosmic abundance ratio of  $^{244}\text{Pu}/^{238}\text{U}$  is revised downwards by a factor of 2.3, from 0.0154 to 0.0068, this revision will have nontrivial implications for some of these problems. A few of these are discussed below:

#### Meteorite Geochronology

Despite its promise,  $^{244}\text{Pu}$  has not been used much as a geochronometer, both because of the difficulty of measurement and lack of a suitable reference element which could be assumed not to fractionate from Pu. Geochronological interpretations based on comparisons on  $^{244}\text{Pu}$  abundances in different samples, e.g. cooling rates based on track densities in different minerals, are not affected by revision in the cosmic abundance estimate. Interpretations based on comparison with assumed cosmic abundance are affected, however. The factor of 2.3 corresponds to 97 Ma. Ambiguities associated with possible chemical fractionation remain as before, of course.

### r-Process Chronology

Many calculations have been performed to examine the relationship between the history of galactic nucleosynthesis and the abundances of specific nuclides in the solar system. Short-lived species, of course, provide the principal constraints on the history of nucleosynthetic activity shortly before the formation of the solar system.

The effect of reducing the value of  $^{244}\text{Pu}/^{238}\text{U}$  has been discussed in detail by Hudson (1981), Jones (1982), and Clayton (1983). Qualitatively, the nature of the modification is clear: downward revision of  $^{244}\text{Pu}/^{238}\text{U}$  requires models which allow for more decay of  $^{244}\text{Pu}$ . This increased amount of decay then predicts less  $^{129}\text{I}$  than is observed to have been present in the early solar system.

Of nucleosynthetic scenarios which could be invoked to resolve this apparent discrepancy, one is obviously suggested by the observations that the even shorter-lived species,  $^{107}\text{Pd}$  (half-life 6 Ma), was also extant in the early solar system. Given that there must have been some relatively freshly synthesized material in the early solar system, it is reasonable to suppose that it included  $^{129}\text{I}$ . Indeed, Kelly and Wasserburg (1978) suggest that because of their proximity in mass, some  $^{129}\text{I}$  must have accompanied  $^{107}\text{Pd}$ . Since  $^{129}\text{I}$  decay in an interval survived by  $^{107}\text{Pd}$  would be minor, this freshly synthesized material would need to account for a fraction of the order of  $10^{-4}$  of the stable and long-lived isotopes; this would have little effect on the  $^{244}\text{Pu}$  abundance. The uncertainties in these calculations are such as to preclude a precise estimate of what fraction of the  $^{129}\text{I}$  was contributed

by the "fresh" material, but it is evidently substantial. Thus the early solar system value of  $^{129}\text{I}/^{127}\text{I} = 10^{-4}$  more nearly reflects mixing of a spectrum of old iodine ( $^{129}\text{I}/^{127}\text{I} \ll 10^{-4}$ ) and young iodine ( $^{129}\text{I}/^{127}\text{I} \gg 10^{-4}$ ) as opposed to a period of unsupported (i.e. no new production) decay of  $^{129}\text{I}$ .

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Table 1. Description of St. Severin Samples

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Label	Weight (g)	Description
SL5	1.034	Whole rock sample of the light lithology irradiated in the SLC5 neutron irradiation
SL	1.555	Aliquot of the SL5 material, not irradiated
SD5	1.075	Whole rock sample of the dark lithology irradiated in the SLC5 neutron irradiation
SD	1.133	Aliquot of the SD5 material, not irradiated
ST	0.221	Hand picked troilite, purity > 99.5%
SW	0.010	Magnetically separated whitlockite, purity 50% with plagioclase as the principle contaminant
SM	0.495	Whole rock sample treated with HNO <sub>3</sub> and magnetically separated to remove much of the plagioclase
SN	0.447	Whole rock sample treated with HNO <sub>3</sub> and magnetically separated to enrich the plagioclase
SF	0.356	Whole rock sample treated with HNO <sub>3</sub> . The fine material from SM and SN not suitable for magnetic separation

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Table 2. Noble gas data for samples of St. Severin

Sample	spallation [ <sup>126</sup> Xe]	trapped [ <sup>130</sup> Xe]	Pu-fission [ <sup>136</sup> Xe]	U-fission [ <sup>136</sup> Xe]	<sup>244</sup> Pu/ <sup>238</sup> U
SL5	0.064	2.3	0.72 ± .09	3.34 ± .07	0.0062 ± .0008
SL	0.066	2.3	0.66 ± .04		
SD5	0.062	3.0	0.78 ± .07	4.40 ± .07	0.0051 ± .0005
SD	0.064	3.0	0.71 ± .04		
SM	0.017	3.0	0.33 ± .04		
SN	0.066	1.8	0.46 ± .04		
SF	0.037	2.8	0.47 ± .04		
ST	0.016	0.5	0.37 ± .04		
SW	2.37	-	61.4 ± .80		

Gas quantities in units of  $10^{-12} \text{cm}^3 \text{STP/g}$