supported by the data. Other possible responses to the 10 January earthquake may be found in 1972 for Fig. 1, b, f, and n.

The third highest energy total for the world occurred in 1969. The largest earthquake in the world for that year and the earthquake responsible for most of the 1969 total occurred off the western coast of Spain on 28 February and had a body wave magnitude of 7.3 (the unusual epicenter of this earthquake is not located within any of the 17 regions investigated). High totals are also found in 1969 for the northern Mid-Atlantic Ridge (Fig. 1b), Africa (Fig. 1e), and the West Indies (Fig. 1p). Since the largest earthquakes in 1969 for all of these regions occurred after 28 February, and since totals for ridge systems are generally higher in 1969 than in 1968, we must again conclude that the second inference is best supported by the data. We note that the most obvious responses occur on ridge systems in close proximity to the magnitude 7.3 earthquake (the northern Mid-Atlantic Ridge and Africa). The other suggested response (the West Indies) also occurs in a region close to this epicenter.

The times of occurrence and epicentral locations of the triggering earthquakes and the triggered earthquakes mentioned in this report imply stress propagation rates on the order of 10^3 to 10^4 km/year. Although no specific rate appears with conspicuous frequency for the events investigated here, there is evidence for global triggering at specific rates in the range 10^3 to 10^4 km/year (*8*).

Chinnery and Landers (*1*) have proposed the existence of worldwide fluctuating stress fields which normally trigger earthquakes only in areas where the ambient stress is very close to that needed for failure. They also suggested that the stresses may occasionally be quite large, resulting in widespread triggering of large events. Anderson (*5, 6*) has proposed that after great decoupling earthquakes, plate motions might be expected to accelerate and trigger earthquakes in adjacent portions of the arc at rates on the order of 10^2 km/year. These hypotheses have been mentioned as possible explanations for unusual periods of global seismic activity. Combining these hypotheses with the limited observations presented here results in an interpretation which could be considered as a refinement of an earlier interpretation (*2, 3*) and as an extension of the proposals of Chinnery and Landers and of Anderson. In this proposed interpretation, major earthquakes are viewed as an immediate cause of fluctuating stress fields which propagate at rates as high as 10^4 km/year, triggering earthquakes on ridge systems where ambient stresses are close to failure.

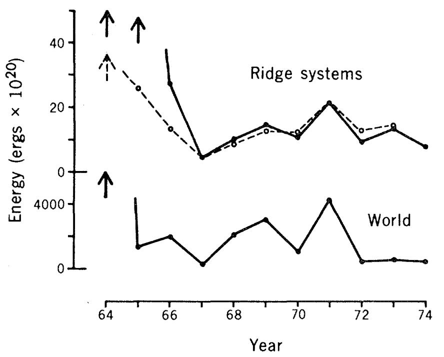

Fig. 2. Yearly seismic energy totals for ridge systems (Fig. 1, a through e) and for the world. The dashed line superimposed on the ridge system totals represents yearly energy totals for the world based on International Seismological Centre data. Arrows indicate energy totals which are off the plots. For more details see (*9*).

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References and Notes

1. M. A. Chinnery and T. E. Landers, *Nature (London)* **258**, 490 (1975).
2. D. A. Walker, *Hawaii Inst. Geophys. Rep. 71-16* (1971). In this study similarities were found in the yearly energy totals for the rising and sinking edges of the Nazca plate. Yearly energy totals before 1966 were not examined.
3. D. A. Walker, G. H. Sutton, G. P. Woollard, N. J. LeTourneau, E. Kausel, *Hawaii Inst. Geophys. Rep. 72-2* (1972).
4. E. Berg and G. H. Sutton, *Phys. Earth Planet. Inter.* **9**, 175 (1974). In this study similarities were found in the cumulative strain release totals for the rising and sinking edges of the Nazca plate. High strain release totals in 1965 were not observed.
5. D. L. Anderson, *Science* **186**, 49 (1974).
6. ______, *ibid.* **187**, 1077 (1975).
7. C. F. Richter, *Elementary Seismology* (Freeman, San Francisco, 1958). The Richter energy-magnitude relation was chosen because of its familiarity. The results of this report are not highly dependent on the formulation assumed.
8. D. A. Walker, in preparation.
9. It has been suggested that NEIS m_b values may be too high because of difficulties in measuring m_b for major earthquakes, and that ISC (International Seismological Centre) m_b values might be more appropriate. Using all earthquakes with ISC m_b values of 6.3 or greater, yearly energy totals for the world have been computed and superimposed on the ridge system totals of Fig. 2. Relative values for the world totals based on the ISC data are similar to those based on NEIS data, while correlations with ridge totals are somewhat better for ISC values than for NEIS values. It should be noted that the total for 1973 is a projected yearly total (141×10^{20} ergs) based on only 6 months of data and that ISC data for 1974 were not available for this investigation. To provide some estimate of scale for the ISC world totals, we also note that the lowest yearly total for the world in 1967 is 42×10^{20} ergs.
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Interrelations Among Isotopically Anomalous Mercury Fractions from Meteorites and Possible Cosmological Inferences

Abstract. *The magnitudes of the mercury anomaly found in unequilibrated meteorites appear to fit a trend. The excesses in the ratios of mercury-202 to mercury-196 are related by simple multiplication factors. This periodicity may be interpreted in terms of the mode of production and ejection of the anomalous isotope from a stellar source.*

Isotopically anomalous $^{202}\text{Hg}/^{196}\text{Hg}$ ratios in a number of unequilibrated chondrites have been reported (*1, 2*). These isotopes were the only ones measured by the neutron activation technique used. The Hg was extracted from irradiated samples of stepwise heating. The isotopically anomalous fractions represent only a very few of the total number of fractions collected in repeated experiments on any particular meteorite. Thus most aliquants contain only material with "normal" Hg—that is, Hg with terrestrial isotopic ratios—or have undergone mixing such that any anomalous Hg is masked because of its low relative concentration. Since experimental procedures could not account for the isotopic variations, it was necessary to suggest a nucleogenetic origin (*1*).

In this report we examine the anomalous fractions themselves in terms of their interrelations, their relation to other fractions released during heating, and some possible implications of these relations. We attempt to recognize trends that will suggest the conditions under which the anomalous isotope was formed and how it became incorporated into material sampled by the solar nebula. The results have cosmological implications, which are discussed.

When the data are examined, it is noted that a number of anomalous Hg isotopic ratios appear to recur. The number of cases with a particular anomalous ratio decreases with increasing magnitude of the anomaly. The ratios are arranged in groups in Table 1 to reflect this tendency. The standard deviations in the means of the groups containing the most entries (groups 1 and 2, containing seven

Table 1. Isotopically anomalous Hg ratios in fractions released from meteorites in stepwise heating experiments; the ratio in normal fractions is taken as 1.00. The ratios are arranged in columns 3 to 7 to reflect the tendency to group in a progressive manner. The total Hg concentration in the samples and the Hg content in the anomalous and the adjacent fractions are given in columns 8 to 11 in parts per billion (ppb). The release temperatures of the anomalous fractions are given in parentheses. Samples in irradiations 18 to 20 are reported in (*1*) and those in 47 to 55 in (*2*). Fractions were collected in 1-hour intervals except where indicated (a to f): a, 17 hours; b, 126 hours; c, 89 hours; d, 2 hours; e, 3 hours; f, 93 hours; and g, 46 hours.

Sample irradiation	Meteorite sample	(Hg concentration from ^{202}Hg)/ (Hg concentration from ^{196}Hg)					Total Hg (ppb)	Hg (ppb) released in anomalous and adjacent temperature steps		
		Group 1	Group 2	Group 3	Group 4	Group 5		Before	Anomalous (°C)	After
54	Murchison bulk	1.18 ± 0.07					> 48	5.47	4.96(600)	1.67
54	Allende matrix	1.17 ± 0.05					32	1.4*	0.14(1200)	
			1.52 ± 0.09					1.94	1.4(600)	0.14*
53	Allende matrix		1.49 ± 0.12				57	2.44	0.79(600)	0.3*
				2.33 ± 0.48				0.79*	0.3(1200)	
52	Allende matrix		1.45 ± 0.11				5.8	0.04	1.45(1200)	
55	Allende matrix < 74 μm	1.17 ± 0.09					241	2.0	2.2(600)	2.5
55	Allende chondrules	1.27 ± 0.09					34	4.3	20(250)	5.6*
		1.20 ± 0.06						20*	5.6a(250)	0.46
47	Allegan chondrules	1.19 ± 0.04					389	107	130(600)	37
			1.40 ± 0.04†					28	25b(250)	16
18	Allegan chondrules					22.8 ± 3.4	1,500	71c	1,120d(300)	4e
18	Orgueil-1	1.25 ± 0.01					16,000	2,700f	209(1200)	
20	Orgueil-2		1.37 ± 0.07				2,500	670g	39(1200)	
20	Hallingeberg			2.28 ± 0.28			470	11.5	2.2(1200)	
19	Tieschitz		1.39 ± 0.06				7,200	56	580(250)	200
					6.1 ± 0.4			92	39(600)	40
Averages		1.20 ± 0.03	1.44 ± 0.05	2.30	6.1	22.8				

*Also an anomalous fraction. †Example of the large initial release of Hg during the first hour of heating at 250°C: first hour, 28 ppb; second hour, 5 ppb; and third hour, 2 ppb; altogether eight fractions were collected in 127 hours (*2*).

and six entries, respectively) are less than 5 percent. Most samples have a ^{202}Hg/^{196}Hg ratio (R) between 1.17 and 1.52. These samples were tested to determine whether they constituted a uniform distribution. The Pearson goodness of fit test indicated that they do not at a .05 significance level. The nonparametric Wilcoxon (Mann-Whitney) (*3*) test of whether the two apparent groups are part of the same distribution indicated that they are not with a significance level of $< .001$. Accepting that these data fall into two groups, the t-test for the difference between two means gave a large difference, hence a very low probability of overlap (significance level $\ll .001$).

The discrete ratios (R) and the number of samples (N) with each ratio are: 1.20 ± 0.03(7), 1.44 ± 0.05(6), 2.30(2), 6.1(1), and 22.8(1). When the ratio in normal fractions, taken to be 1.00, is subtracted from these, the values of $\Delta = (R - 1)$ for the groups are: 0.20, 0.44, 1.30, 5.1, and 22. This is a progression which can be generated, using 0.20 as base, by $0.20 \times N!$.

In order to determine whether the occurrence of the anomalous ratios in such an ordered manner is significant we must examine the Hg release data.

Samples were generally collected at 100° or 130°, 150° or 175°, 250° or 300°, 450°, 600°, and 1200°C. The temperature step during which the anomalous fraction was released is indicated for each entry in Table 1. This fraction usually appeared at one of three temperatures: 250°, 600°, or 1200°C; some samples released anomalous Hg at two of these temperatures. Bulk samples usually released anomalous Hg at the higher temperatures; chondrule samples tended to release anomalous Hg at the lower temperatures. The low-temperature release may be due to posttrapping effects such as devitrification of the glass in chondrules. There is no relation between the temperature of collection and the magnitude of the isotopic anomaly, when present.

To evaluate possible dilution (interstep contamination) effects, not only the isotope ratio in the adjacent fractions but also the concentrations of Hg released are examined. The concentrations in the anomalous fraction and in the "before" and "after" fractions are listed in Table 1, columns 9 to 11. Three cases are apparent.

1) The most unambiguous case is that in which either the before or the anomalous fraction was held at a given temperature for times greater than 1 hour (indicated in Table 1), especially where more than one sample was collected during this protracted heating. Allende chondrules (irradiation 55), Allegan chondrules (irradiation 47), Tieschitz at 250°C, and Orgueil-1 and Orgueil-2 at 1200°C fall in this category.

2) The second most definitive situation is when the before fraction contains less Hg than the anomalous fraction, so that an Hg residual from the before temperature step will have only a small effect, for example, Allende matrix (irradiation 52).

3) When the Hg concentration in the before fraction is equal or greater than that in the anomalous fraction, only a small dilution of the latter is expected, since experience shows that the largest release is during the first hour of heating. See the data for Allegan chondrules (irradiation 47) in Table 1. One may also resort to circular reasoning and argue that since all the data are consistent with the systematics noted earlier—that is, belong to one or another of the ratio groups—then dilution effects have been negligible.

We noted that the anomalous ratios are not correlated with temperature; it can be seen that they are not correlated with concentration or fraction of total Hg either.

These quantized ratios impose the following constraints on the source of the anomalous Hg: (i) the Hg reservoir sampled by each group was discrete but related to that sampled by other groups; (ii) the phase or phases that incorporated the anomalous fractions were unique in that similar material did not incorporate the usually dominant normal Hg; and (iii) random mixing did not occur between Hg atoms in sites with different isotope ratios. The inference is that the sources and conditions were different for the incorporation of the anomalous and normal Hg. It appears that the anomalous

Hg itself, including that in chondrules, was trapped exclusively at high temperatures ($\geq$ 600°C).

We interpret these observations as follows.

1) If a single source (star) produced all the Hg then we have sampled this source either at various stages in its evolution (one stage in which normal Hg was synthesized, another in which anomalous Hg was being or had been synthesized) or at a stage in which neutron-rich isotopes were synthesized, followed by synthesis of neutron-deficient ^{196}Hg. The source was periodically sampled, possibly by ejection. Each ejection sampled a different substage in the process that was related to a relative underproduction of ^{196}Hg or overproduction of ^{202}Hg.

2) If multiple sources (stars) are invoked, only two types are allowed. One object or class of objects produced normal Hg; the other had to be a single object in order to produce anomalous Hg in a progressively varying manner.

3) Anomalous Hg produced by a single event in a star became mixed in a quantized way with normal Hg from the same or some other star.

In case 3 a mechanism must be invented to provide batchwise and correlated mixing of normal and anomalous Hg and then fix each mixture in a carrier to the exclusion of most of the normal Hg in the system. In contrast, cases 1 and 2 allow mechanisms for the production and trapping of the anomalous component but may present problems in the identification of the sources. We have not found ways of achieving case 3, but propose models for cases 1 and 2. In these cases the periodicity of the Hg isotope ratios may be interpreted as time-related. A process occurred which involved a periodic ejection of material that was decaying or being generated in the source. Each ^{202}Hg/^{196}Hg group, then, sampled the source at specific times determined by the source, and the material sampled was at a different stage of evolution—destruction (decay) or production—in the source. In a cooling gas cloud, for example, we might expect a number of trapping sites to become available. Since the major-element compositions of a star probably do not change greatly each time a stellar envelope develops and is ejected, the type of trapping site should be repeated.

We may examine two possible processes.

1) A process that produced ^{202}Pb had occurred and terminated before the star began to explosively eject material. The ^{202}Pb decayed to ^{202}Hg with a half-life of $\sim 3 \times 10^5$ years. This process is intriguing since it permits assigning a time scale to a phase of stellar evolution. Since the ratio ^{202}Hg/^{196}Hg = 22.8 indicates that there is $\sim$ 95 percent ^{202}Hg in the sample (considering only ^{196}Hg and ^{202}Hg), the time from the end of ^{202}Pb production to the initiation of pulsing is $\sim 2.2 \times 10^4$ years. The assumption is that the ^{202}Pb that gave rise to the apparent excess in ^{202}Hg was equally efficiently trapped, but in different sites than Hg, each time it was ejected from the star. The time between pulses decreased in steps: $6.97 \times 10^5 \rightarrow 6.00 \times 10^5 \rightarrow 4.76 \times 10^5 \rightarrow 3 \times 10^5$ years (*4*). Thus, in $\sim 2.1 \times 10^6$ years the ^{202}Pb would have decayed to a level below our sensitivity. It should be noted that the ^{202}Hg resulting from ^{202}Pb decay in the source would be masked by normal Hg and also trapped as Hg and thus would not be detectable.

Table 2. Concentrations of Hg released at 250°C based on the mass 196 and mass 202 isotopes. Asterisks indicate that the samples were anomalous; extraction times are given in parentheses.

Hg (ppb)	Irradiation			
	21-1 (66 hr)	21-2 (18 hr)	47* (126 hr)	18* (2 hr)
From mass 196	75	256	46	50
From mass 202	75	256	53	1120

2) A process began in which ^{196}Hg production started. This had to be at the termination of a rapid (r) or slow (s) neutron capture process which produced ^{202}Hg and other neutron-rich Hg isotopes and the beginning of a proton (p) process to produce ^{196}Hg. The source of the ^{196}Hg had to be isolated from the source of normal Hg, otherwise the expected abundant normal Hg would have masked the Hg depleted in mass 196. As ^{196}Hg was being produced in the star, each pulse ejected Hg with an increasing complement of ^{196}Hg until the relative concentration of ^{196}Hg approached that found in normal Hg. The process must have ceased about this time since we have not observed a sample with excess ^{196}Hg.

Results of some experimental studies of meteorites are pertinent to the case ^{202}Pb $\rightarrow$ ^{202}Hg. There are two types of possibly negative and two types of possibly supportive evidence.

Preliminary mass spectrometric data on Hg volatilized from Allegan chondrules in a manifold in a 100-inch mass spectrometer by Stevens (*5*) indicates normal heavy isotopic abundances and depleted ^{196}Hg. This result must be confirmed.

The isotopic composition of Tl in meteorites measured (with greater precision than our Hg ratios) by Anders and Stevens (*6*) and by Ostic *et al.* (*7*) is normal. Thallium-205 has a possible parent in ^{205}Pb (half-life, 3×10^7 years). The usually accepted mode for making ^{205}Pb is the s-process. However, if ^{202}Pb was made, then presumably ^{205}Pb could have been produced by some of the same types of nuclear reactions. Failure to detect an effect could occur if dilution by normal Tl masked any isotopic variations, or it simply could be due to sampling.

The experimental support for the anomaly being in the mass-202 isotope is found in the four sets of Allegan chondrule samples we have measured. Two Allegan chondrule samples released anomalous Hg; the other two showed no anomaly. The Hg contents in the 250°C fraction from the four sets of Allegan chondrules are given in Table 2. It seems from the very large concentration of Hg based on mass 202 in sample 18 that the ^{202}Hg isotope is anomalous.

An isothermal diffusion experiment at 250°C on Allegan chondrules (irradiation 47) established that the two isotopes were released with different activation energies—that is, at different rates or from different sites in the phase containing the Hg (*2*). Assuming that ^{202}Pb had a role in producing excess ^{202}Hg might explain this difference in the activation energy. If devitrification of the chondrule glass occurred before ^{202}Pb decay then the two Hg isotopes could become concentrated in different sites.

We comment briefly on the O and Mg isotopic anomalies in high-temperature inclusions in type-2 and type-3 carbonaceous chondrites (*8*) reported by Clayton *et al.* (*9*) and by Gray and Compston (*10*), Lee and Papanastassiou (*11*); and Lee *et al.* (*12*), respectively. An interpretation similar to that proposed for anomalous Hg may be considered. Both the ^{26}Mg and the ^{17}O and ^{18}O data appear to form discrete groupings. There is a periodicity in these groupings: relative enrichments in ^{26}Mg differ by multiples of $\sim$ 1.5 to 1.6 and relative depletions in ^{17}O and ^{18}O differ by a similar factor. An additional point of interest is that the ^{26}Mg excess has been attributed to the decay of radioactive ^{26}Al (*10–14*). We suggest that there may be a relationship among the Hg, Mg, and O results. The nucleogenetic picture is complex. A possibility is that the p-reaction, whether proton or photon, that may have made ^{26}Al ($\rightarrow$^{26}Mg) and either ^{202}Pb ($\rightarrow$^{202}Hg) or ^{196}Hg may have effectively destroyed ^{17}O and ^{18}O, leading to highly depleted O. Regardless of the nuclear process, there must have been very little mixing of the materials ejected at different times (*4*, *14*). These materials did not exchange isotopically with more abundant normal material, either before the formation of the solar nebula, in the solar nebula, or in the meteorite parent body. The Hg data indicate that only a low-temperature (< 100°C) history is acceptable for much of this matter. Suggestions for proton types of processes have been made by Heymann and Dziczkaniec (*13*) and by Turkevich (*15*). In our model these reactions would occur in a presolar nebula source.

Finally, the Hg results may be considered, to some extent, to be consistent with the proposal by Clayton (*16*) that anomalous Xe or its progenitor was trapped in circumstellar grains associated with explosive stars. We go beyond this proposal in requiring that the star eject material periodically.

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References and Notes

1. G. W. Reed and S. Jovanovic, *J. Inorg. Nucl. Chem.* **31**, 3783 (1969).
2. S. Jovanovic and G. W. Reed, Jr., *Earth Planet. Sci. Lett.* **31**, 95 (1976).
3. D. B. Owen, *Handbook of Statistical Tables* (Addison-Wesley, Reading, Mass., 1962), test 11.1.
4. The time intervals between pulses, Δt, are determined by the fractional ^{202}Pb decay needed to give the observed excess of ^{202}Hg in each step. Thus, $\Delta t = (\ln 2/T)\ln(A_j/A_1)$ where the half-life, T, for ^{202}Pb is ~ 3 × 10^5 years and A_j/A_1 = 5.1/22.8, 1.3/5.1, 0.44/1.3, and 0.20/0.44 for the fractional anomalies ($A_j, A_1, \ldots$) we observe.
5. C. M. Stevens, personal communication.
6. E. Anders and C. M. Stevens, *J. Geophys. Res.* **65**, 3043 (1960).
7. R. G. Ostic, H. M. El-Badry, T. P. Kohman, *Earth Planet. Sci. Lett.* **7**, 72 (1969).
8. We determined Hg in high-temperature inclusions but found no anomaly in the fractions containing enough Hg for accurate measurements (*2*).
9. R. W. Clayton, L. Grossman, T. K. Mayeda, *Science* **182**, 485 (1973).
10. C. M. Gray and W. Compston, *Nature (London)* **251**, 495 (1974).
11. T. Lee and D. A. Papanastassiou, *Geophys. Res. Lett.* **1**, 225 (1974).
12. ———, G. J. Wasserburg, preprint.
13. D. Heymann and M. Dziczkaniec, *Science* **191**, 79 (1975).
14. If periodic ejection from a star accounts for the ^{26}Mg anomaly, the periods may be calculated by using the ^{26}Al half-life of 7.4 × 10^5 years. The intervals calculated are somewhat longer than those based on ^{202}Pb. The pulsing intervals calculated from ^{26}Al and ^{202}Pb decay will coincide if the ^{202}Pb half-life is 2.4 × 10^5 years rather than the ~ 3 × 10^5 years used in the text.
15. A. L. Turkevich, paper presented at the American Geophysical Union Meeting, Washington, D.C., April 1976.
16. D. D. Clayton, *Astrophys. J.* **199**, 765 (1975).
17. We especially wish to thank A. H. Jaffey for his assistance with statistical treatment of the data. Fruitful discussions with a number of colleagues, especially C. M. Stevens, M. S. Freedman, and P. A. Benioff, are gratefully acknowledged. We thank D. Schramm and B. Perry for their thoughtful comments. D. Heymann's thorough and critical review of our original manuscript led to major revision and improvements; however, the authors are solely responsible for this presentation.

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Binding of [^{14}C]Parathion in Soil: A Reassessment of Pesticide Persistence

Abstract. *A steady decrease of extractable [^{14}C]parathion residues in soils over a 1-month incubation period was accompanied by an increase of unextractable, bound ^{14}C-labeled residues, resulting finally in total recoveries of extracted plus bound residues of 80 to 87 percent of the applied radiocarbon. Soils containing bound residues were nontoxic to fruit flies. Binding of ^{14}C-labeled residues was related to the activity of soil microorganisms; soil sterilization resulted in a reduction of binding by 58 to 84 percent. Under flooded (anaerobic) conditions, the binding of compounds labeled with ^{14}C doubled, and parathion was reduced to aminoparathion. Reinoculation of sterilized flooded soil fully reinstated the binding capacity. [^{14}C]Aminoparathion was preferentially bound to soil, since its binding within 2 hours was 30 times greater than that of [^{14}C]parathion. Because of the existence of formerly "unseen," unextractable residues, the concept of "persistent" and "nonpersistent" pesticide residues might have to be reconsidered.*

Difficult problems have emerged regarding the ultimate fate of synthetic chemicals for agricultural pest control. Before 1962 both analytical methodology and knowledge about the metabolism of many pesticides were limited, so that analyses for these compounds were primarily for the applied "parent" compound. In addition, only easily extracted residues were detected. Accordingly, typical depletion curves for several chlorinated hydrocarbon insecticides in soils were published by this laboratory, showing the "persistence," "disappearance," or "loss" of the pesticides (*1*, *2*). The relatively short persistence of parathion residues under both laboratory and field conditions was described a few years later (*3*). When the fate of pesticide residues was described, the vague terms "disappearance" or "volatilization" were widely used, but they do not include the formation of unextractable bound residues. The introduction of radiolabeled pesticides for experimental use made it possible to obtain a "balance" and to account for the fate of the applied radiocarbon. The use of combustion or strong hydrolytic techniques with the extracted soils revealed that unextracted or bound residues could be released and detected, as for example, with the herbicide propanil (*4*) and the insecticide dyfonate (*5*). The problem of the bound residues, however, is complicated since present methods for their release (combustion, hydrolysis) also result in the destruction of their identity.

Earlier we investigated the persistence of the organophosphorus insecticide, parathion, in a loam soil (*3*); we now report a study conducted with the same soil and an additional sandy soil, using parathion (*O,O*-diethyl *O*-*p*-nitrophenyl phosphorothioate) labeled with ^{14}C either in the aromatic ring or in the ethoxy group. In this way, we intended to quantitatively ascertain the fate of [^{14}C]parathion in an attempt to clarify some questions pertaining to the mechanism of its binding to soils.

Preliminary studies were carried out with samples of a sandy soil collected periodically from a Wisconsin cranberry bog where parathion is used extensively, and the soil is periodically flooded and drained. In the laboratory, soil samples were treated with parathion labeled with ^{14}C in the ring ([*phenyl*-^{14}C]parathion) at a concentration of 1 to 2 parts per million (ppm) and incubated under moist conditions at 23°C in the dark. After 14 days the soil was extracted three times by a routine method with a mixture of benzene, methanol, and acetone (1 : 1 : 1). A portion was then examined for the presence of unextractable or bound ^{14}C-labeled residues by burning it to $^{14}CO_2$ in a Packard model 305 Tri-Carb sample oxidizer (*6*). Up to 60 percent of the originally applied radiocarbon (in the form of [^{14}C]parathion) was recovered in the form of bound residues. In an attempt to release the bound residues, samples of these soils were further extracted by using various solvent mixtures that ranged in polarity from benzene to water. The total ^{14}C recovered by six additional extractions amounted to only 2.4 percent of the applied radiocarbon, while 56.5 percent still remained in the soil in unextractable bound residues. Autoclaving and subsequent extraction of this soil with a mixture of benzene, methanol, and acetone (1 : 1 : 1) and then with ethyl acetate resulted in an additional release of only 1.2 percent of the applied radiocarbon.

After these preliminary findings, detailed experiments were carried out with a sandy soil from a cranberry bog [composition in percentages: organic matter, 1.3; sand, 98; silt, 2: and clay, 0 (*p*H 7.2)] and Plano silt loam soil [composition in percentages: organic matter 4.7; sand, 5; silt, 71; and clay, 24 (*p*H 6.0)].