

THE EXTRATERRESTRIAL COMPONENT IN MARINE SEDIMENTS: DESCRIPTION AND INTERPRETATION

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Abstract. The influx of extraterrestrial matter to the Earth is dominated by two size-fractions: sub-millimeter interplanetary dust and impacting asteroids and comets. Over geologic time the major contribution of extraterrestrial matter is from the largest impactors. Interplanetary dust is largely vaporized by atmospheric entry, but some surviving material is found in sediments as cosmic spherules. Impacting asteroids and comets can produce fallout layers either regionally or globally which may contain a significant fraction of meteoritic material as well as shock-metamorphosed and shock-melted terrestrial material. Iridium is a sensitive tracer of extraterrestrial matter. Although high concentrations of Ir in marine sediments probably have an extraterrestrial origin, they do not necessarily indicate the presence of a major impact event. Of the thousands of impact horizons which must be resolvable in the sedimentary record, to date only seven probable impact horizons have been identified in the entire Phanerozoic. The marine geochemistry and occurrence of Ir is still poorly understood, but the thousands of Ir analyses performed in the last several years have demonstrated that the global occurrence high Ir (>10 ng/g) concentrations in Cretaceous-Tertiary boundary sediments is a truly anomalous phenomenon.

INTRODUCTION

The study of extraterrestrial matter in marine sediments began over a century ago with the discovery by Murray and Renard [1891] of cosmic spherules in the magnetic fraction of red clays dredged on the HMS *Challenger* expedition. In the succeeding decades a variety of techniques have been employed

to estimate the amount of the extraterrestrial component. The technique which has proven the most sensitive tracer of extraterrestrial matter is the Ir concentration of marine sediments. Since Ir is depleted in continental crust by about 10^4 relative to chondritic meteorites, high concentrations of Ir in marine sediments probably have an extraterrestrial origin.

The discovery of anomalous concentrations of Ir in Cretaceous-Tertiary boundary sediments [Alvarez et al., 1980] and the resultant hypothesis that this boundary recorded the impact of a 10-km asteroid or comet has generated considerable interest in the general scientific community on the record of extraterrestrial matter in sediments. This interest has been both in the possibility that major impact events may be responsible for worldwide ecological crises and in demonstrating whether high Ir contents consistently reflect accretionary events.

This paper is meant to be a general (although not comprehensive) review of the occurrence of extraterrestrial matter in marine sediments. Topics discussed include (1) the mass influx rates of interplanetary dust and impacting asteroids and comets, (2) the types of deposits they might be expected to produce, (3) the use of Ir as a tracer of extraterrestrial matter, (4) a list of probable and possible impact horizons in the geological record, (5) the marine geochemistry of Ir, (6) the comparison of a 34-Ma Ir profile to that of four other trace elements, (7) periodic comets showers, and (8) Re-Os isotopic systematics of marine sediments.

EXTRATERRESTRIAL MASS INFLUX RATES

Extraterrestrial matter is accreted to the Earth in a broad range of sizes from microscopic dust to mountain-sized asteroids and comets. This influx is illustrated in Figure 1 which plots the global mass influx per year for each magnitude of mass between 10^{-14} and 10^{18} g. One can see that there are two dominant sources of extraterrestrial matter: the interplanetary dust complex and large asteroids and comets.

On relatively short time scales ($<10^4$ years) the background

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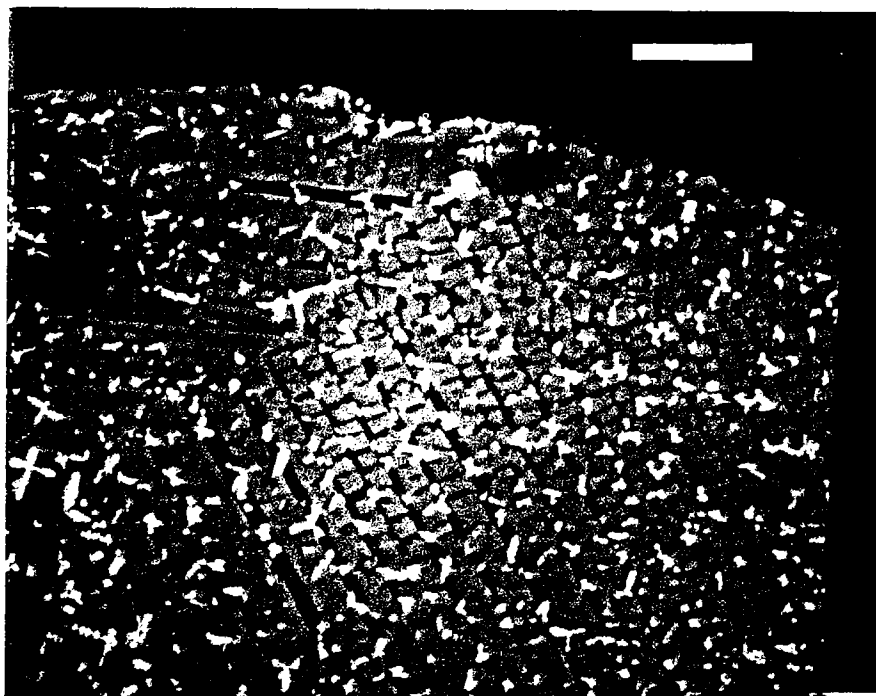


Fig. 2. Backscatter electron image of a portion of a polished section of a stony cosmic spherule composed of olivine and magnetite (small bright grains). The interstitial voids were originally filled with glass which has been dissolved during alteration. The small very bright grain at the top of the spherule is a Pt-metal nugget containing Ru, Rh, Pd, Os, Ir, Pt, and Au (scale 10 μm).

solar wind-implanted He [Rajan et al., 1977]. The elevated ^3He concentrations in sediments suggest that these particles also survive long enough to be buried.

The sub-millimeter particles which dominate the mass of the interplanetary dust complex have a very low probability of surviving atmospheric entry. Bates [1986] has modeled the atmospheric entry of 50- to 500- μm interplanetary grains. Although large particles with low entry angles (5° - 10°) and low velocities ($<15 \text{ km s}^{-1}$) can survive as intact micrometeoroids, in the typical case, $>90\%$ of total mass is lost by vaporization. The surviving material is in the form of quenched molten droplets known as cosmic spherules. Cosmic spherules are recovered from the magnetic fraction of slowly accumulating marine sediments. They occur as two main types: stony and iron [Blanchard et al., 1980, Brownlee, 1981]. The stony spherules have an ultramafic, chondritic composition and are characterized by a mineral assemblage of olivine, magnetite, and glass (Figure 2). Unmelted 'relict' mineral grains from the meteoritic precursor are occasionally found in stony spherules. These spherules are chemically unstable and have only been discovered in young surface sediments. The iron-type cosmic spherules typically contain iron oxides (magnetite and/or wustite) and may contain a metallic core. Nickel may occur in the oxides and is invariably present in the metallic cores. They are more resistant to alteration than stony spherules. Although metal cores are unstable, the oxide shells can survive millions of years. Iron-type cosmic spherules have

been found in Paleozoic salts [Mutch, 1964, 1966], Jurassic cherts [Mutch and Garrison, 1967], and Jurassic hardgrounds [Czajkowski, 1987].

Brownlee et al. [1984] discovered that a large fraction of iron-type cosmic spherules contain small ($<10 \mu\text{m}$) nuggets of Pt-group metals. More recently, Bonte et al. [1987] have shown that Pt-metal nuggets are also common in stony spherules. They found that nearly all of the Ir (the Pt-metal they analyzed) in 16 iron and 8 stony cosmic spherules is contained in discrete, micrometer-sized nuggets (Figure 2). These nuggets are extremely stable and corrosion resistant in most sedimentary environments. The discovery of Pt-metal nuggets raises the intriguing possibility that most of the Pt-metals from melted interplanetary dust enters these grains. Because they are so insoluble, chemical analyses of Pt metals (such as Ir) in sediments should employ sample dissolution techniques that will dissolve these nuggets.

Asteroidal and Cometary Impactors

The accretion of large fragments of asteroids and comets results in a geologically instantaneous input of a large amount of extraterrestrial matter. In marine sediments the record of this type of event will be a sedimentary horizon enriched in meteoritic and possibly impact-derived terrestrial material. The specifics of the impact and dispersal of this material is poorly

now recognized as a global phenomenon [e.g., Alvarez et al., 1984]. Ir-rich KT sediments occur in both marine and nonmarine [Orth et al., 1981] sedimentary sections. Impact debris in the form of submillimeter shocked minerals [Bohor et al., 1984, 1987] and spherules with high-temperature spinels [Smit and Kyte, 1984; Kyte and Smit, 1986] has been found in several localities. Although some workers [e.g., Officer and Drake, 1985] argue for a volcanic source for the high concentrations of Ir and other noble metals in KT sediments, this must be considered a low-probability alternative at present. Admittedly, proponents of the volcanic alternative would disagree with this assessment. However, this author agrees with Alvarez [1987] that the bulk of the available data is not only consistent with an impact event, it fulfills many predictions of the impact hypothesis. On the other hand, there is no compelling evidence to suggest that the KT boundary clay could be derived from volcanism. In order to explain the global occurrence of shocked quartz [Bohor et al., 1987], Officer et al. [1987] proposed that this could be accomplished by massive amounts ($\sim 3 \times 10^6$ km³ of magma) of explosive (presumably silicic) volcanism. A silicic ash would have distinctive chemical, isotopic, and mineralogical characteristics. Izett and Bohor [1987] have pointed out that KT boundary clays do not contain a coherent assemblage of euhedral volcanic minerals (e.g., bipyramidal quartz, zircon). Certainly, if KT boundary clays are volcanic deposits, one should be able to prove this with more definitive evidence than high concentrations of Ir and shocked minerals.

Although high concentrations of Ir in marine sediments can be an indication of high concentrations of meteoritic material, Ir analyses should be supplemented by mineralogic, stratigraphic, and sedimentologic analyses to confirm the nature of the signal. High concentrations of meteoritic Ir may result from impacts, low sediment accumulation rates, or long periods of nondeposition. An impact origin may be indicated by Ir accumulation rates considerably higher than expected within a given stratigraphic interval. The occurrence of high-pressure and high-temperature mineral phases should also be expected. Although high concentrations of Ir are likely to reflect a large extraterrestrial component, this is not in itself sufficient to indicate an impact event. An excellent example is the study of three Jurassic hardgrounds by Czajkowski [1987] which demonstrated that long periods of nondeposition can result in lag deposits rich in cosmic spherules and Ir. In one hardground, Czajkowski measured an Ir concentration of 20 ng/g, more than two orders of magnitude higher than that expected for typical marine sediments.

PROBABLE AND POSSIBLE IMPACT HORIZONS IN THE SEDIMENTARY RECORD

The discovery of the KT Ir anomaly has resulted in considerable interest in a possible connection between impact events and extinctions in the Phanerozoic. Several researchers have measured the Ir concentrations in numerous samples of geologic boundaries. This research has had mixed success, with several positive, negative, and conflicting results. The horizons that can most probably be attributed to impacts are in the Cenozoic (except one in the Precambrian), and all of these are supported by mineralogical evidence independent of Ir

TABLE 1. Probable Impact Horizons With Ir or Impact Debris

| Horizons | Age, Ma | Remarks |
|--------------------------------------|---------|----------------------------|
| 1. Microtektite Horizons: | | |
| Australian-Indonesian | 0.71 | no Ir |
| Ivory Coast | 1.15 | no Ir |
| North American | 38 | no Ir |
| 2. Clinopyroxene-bearing spherules | 38 | low Ir (2 horizons?) |
| 3. Late Pliocene (southeast Pacific) | 2.3 | high Ir regional? |
| 4. Cretaceous-Tertiary Boundary | 66 | high Ir global |
| 5. Late Precambrian | 600 | shocked rocks and minerals |

concentrations. Although there have been several reports of high-Ir layers in Paleozoic and Mesozoic sediments, their interpretation as impact events is presently tenuous.

Probable Impact Horizons

Table 1 is a list of probable impact horizons in the sedimentary record. These are listed as probable on the basis of mineralogical evidence independent of high Ir concentrations. The Late Pliocene event has been identified in two piston cores in the southeast Pacific and contains high concentrations of Ir and Au [Kyte et al., 1981], as well as impact melt debris and unmelted meteoritic material [Kyte and Brownlee, 1985]. Present estimates of the size of the late Pliocene projectile are on the order of 0.5 km [Kyte et al., 1987]. The microtektite horizons in the Pleistocene and Eocene are correlated to tektite-strewn fields and are believed to be glasses quenched from impact melt droplets [Glass et al., 1979]. The recent discovery of coesite in the late Eocene horizon [Glass, 1987] confirms its impact origin. An interesting point is that there are no Ir anomalies associated with the microtektite horizons. Either these were produced by the impact of nonchondritic asteroids having low concentrations of Ir, or the ejecta in these horizons have extremely low projectile/target ratios.

Recently, Glass et al. [1985] recognized a late Eocene horizon containing clinopyroxene-bearing (cpx) spherules that is distinct from the North American microtektite horizon. The cpx-spherule horizon was initially associated with the North American tektite field and microtektites; the discovery that this was a separate horizon was mainly the result of Ir analyses which demonstrated that anomalous concentrations of Ir were stratigraphically below the North American horizon [Ganapathy, 1982; Sanfilippo et al., 1985]. Recently, Keller et al. [1987] reported that the cpx-spherule horizon may represent two separate events in the Late Eocene, but this interpretation has been disputed by Glass and Burns [1987]. At most

TABLE 3. Chemically Investigated Geologic Boundary Sections With No Evidence of an Impact

| Boundary | Locality | Age, Ma | Reference |
|-----------------------------|----------------------------------|---------|---|
| Cenomanian-Turonian | England | 91 | Orth et al. [1985] |
| Pliensbachian-Toarcian | England | 193 | Orth et al. [1985] |
| Triassic-Jurassic | Alaska | 208 | Orth et al. [1985] |
| Permian-Triassic | China " USSR | 245 | Clark et al. [1986] Zhou and Kyte [1986] Aleksiev et al. [1983] |
| Mississippian-Pennsylvanian | Oklahoma Texas | 320 | Orth et al. [1986c] " |
| Devonian-Mississippian | Oklahoma | 360 | Orth et al. [1986a] |
| Frasnian-Famennian | Belgium New York Germany | 367 | McGhee et al. [1984] " McGhee et al. [1986] |
| Ordovician-Silurian | Scotland Quebec | 428 | Wilde et al. [1986] Orth et al. [1986b] |
| Cambrian-Ordovician | England Canada Scandinavia | 505 | Orth et al. [1985] " " |
| Cambrian-Biomeres | Utah | 520 | Orth et al. [1984] |
| Precambrian-Cambrian | USSR | 570 | Orth et al. [1985] |

states to tetravalent ones during mineral formation. Goldberg et al. [1986] suggest that Pt and Ir are mainly precipitated in hydrogenous Mn oxides and that the seawater Pt/Ir ratio is found only in nearly pure hydrogenous ferromanganese deposits.

The long residence time of Ir (relative to the mixing time of the oceans) and its tendency to precipitate in hydrogenous minerals should result in a relatively uniform deposition rate in marine sediments. Since the influx of interplanetary dust, as well as asteroids and comets (with due allowance for stochastic effects) is also expected to be relatively constant, this should also result in a relatively uniform accumulation of Ir-bearing particulates (note that interplanetary dust and large projectiles should also contribute to the dissolved Ir as a result of vaporization of dust in the atmosphere and shock vaporization of a large amount of impacting projectiles). Thus the background accumulation of Ir both as extraterrestrial

particulates and hydrogenous precipitates should combine to produce concentrations inversely proportional to the sedimentation rate as noted by Barker and Anders [1968]. However, high Ir accumulation rates over geologically brief intervals probably represent rapid accretion of large amounts of extraterrestrial material (e.g., a major impact event).

The most thorough analysis of Ir in a single sedimentary section was by Kyte and Wasson [1986a]. They reported analyses of Ir in 149 samples across a continuous 9-m section of abyssal clay (piston core LL44 GPC3) from the north central Pacific. Their stratigraphic control was based on ichthyolith biostratigraphy [Doyle, 1980] which indicated that the section encompassed the time from the KT boundary to the early Oligocene. These Ir data (Figure 3) confirmed two important hypotheses: (1) the highly anomalous character of the KT boundary and (2) the relatively constant background accumulation rate of Ir in marine sediments as proposed by Barker and Anders [1968]. Preliminary Ir data for an additional 12 m of this section (F.T. Kyte, unpublished data, 1988) confirm that the KT boundary Ir anomaly is unique in the Cenozoic, at least within the constraints available in the stratigraphy of this clay section. These data, along with profiles of 25 other trace elements, will be published elsewhere. Kyte and Wasson [1986a] found that the Ir accumulation rate in the ~30 Ma of section above the KT boundary was $13 \text{ ng cm}^{-2} \text{ Ma}^{-1}$. They suggested that the contribution of extraterrestrial Ir was $9 \pm 3 \text{ ng cm}^{-2} \text{ Ma}^{-1}$. This corresponds to an annual accretion rate of 78 Gg a^{-1} of extraterrestrial matter, a value consistent with the estimated influx of objects ranging in mass from 10^{14} to 10^{17} or 10^{18} g. Unfortunately, the Ir background concentrations in this section were too high to resolve events much smaller than the KT; to date, the Late Eocene cpx-bearing spherule horizon has not been resolved, although there are hints of its presence.

Kyte and Wasson [1986a] suggested that the Ir profile in LL44 GPC3 indicates that this element behaves like the largely hydrogenous element Co. This is the behavior predicted for dissolved Ir by Goldberg et al. [1986] and by the assumption of a relatively constant accumulation rate of meteoritic particulates. For comparison, concentration versus depth profiles are given for Co, La, Sb, and Cr (Figure 3). Each has a different abundance profile, reflecting the relative importance of different sedimentary components in LL44 GPC3. Identification of these components is preliminary at present and is based largely on comparison with interpretations of earlier chemical analyses and partitioning experiments on this section by Leinen [1987]. Cobalt is probably largely contained in hydrogenous Mn oxides and may reflect a relatively constant precipitation rate from seawater superimposed on variable sediment accumulation rates. The plateau of high Ir and hydrogenous Co from the late Paleocene to early Oligocene are consistent with low sediment accumulation rates at this time [Doyle, 1980]. The profile for La is typical of the rare earth elements, and the large peak from ~16 to 19 m correlates well with high concentrations of phosphatic fish debris observed in this part of the section by Doyle [1980]. The Sb profile displays two large peaks in the Eocene which might reflect enhanced accumulation of a hydrothermal component derived from the East Pacific Rise. Leinen [1987] estimated that the hydrothermal component in this part of the section may be as

extinction events at the KT and Eocene-Oligocene boundaries were caused by periodic showers of comets.

Periodic comet showers have been invoked by several researchers as an astronomical mechanism capable of generating the periodicity in terrestrial extinction rates suggested by Raup and Sepkoski [1984]. The hypothetical comet showers are caused by perturbation of the Oort cloud (a spherically symmetric cloud of comets extending out to interstellar distances [e.g., Weissman, 1985]) by (1) a dark companion star to the sun [Whitmire and Jackson, 1984; Davis et al., 1984], (2) giant molecular clouds encountered during the Earth's oscillation through the galactic plane [Rampino and Stothers, 1984] or (3) by an unobserved tenth planet [Whitmire and Matese, 1985].

Showers of $\sim 2 \times 10^9$ new long-period comets (as suggested by Davis et al. [1984]) would only occasionally cause impacts as large as that hypothesized for the KT boundary [Kyte and Wasson, 1986b]. However, they would result in an increase in the long-period comet flux by factors of at least 200 and should cause the total Ir accumulation rate to increase by at least an order of magnitude over a period of 1–3 Ma [Kyte and Wasson, 1986a]. Large reductions in the hypothesized size of the comet showers as suggested by Muller [1986] would avoid this constraint but would also reduce the impact rate to levels insignificant relative to the random background [Kyte and Wasson, 1986b]. Unless a significant shift in the background Ir accumulation rate at the times proposed for these showers can be documented, their existence should be considered highly questionable.

RE-OS ISOTOPIC SYSTEMATICS IN MARINE SEDIMENTS

An important new advance in measuring the extraterrestrial component in marine sediments has been in the application of Re-Os isotopic systematics (for a summary of applications of the Re-Os system, see Faure [1986]). Both Re and Os are siderophile elements which are depleted in the Earth's mantle relative to chondrites. However, unlike Ir and Os, Re is fractionated into the crust during magmatism, and crustal Re concentrations are slightly greater than in mantle peridotites. The crustal Re/Os ratio is about 100 times that in chondritic meteorites or the mantle.

Because ^{187}Re decays to ^{187}Os ($t_{1/2}$, 4.27×10^{10} years), the high Re/Os ratio in the crust has resulted in relatively high crustal abundances of the ^{187}Os isotope (this is typically reported as a ratio of $^{187}\text{Os}/^{186}\text{Os}$; ^{186}Os is nonradiogenic). The $^{187}\text{Os}/^{186}\text{Os}$ ratio in meteorites and the mantle is ~ 1.1 while the $^{187}\text{Os}/^{186}\text{Os}$ ratio in continental crust must be between 13 and 30, depending on the model of crustal evolution used [Turekian, 1982]. Thus the $^{187}\text{Os}/^{186}\text{Os}$ ratio has the capability of distinguishing between continental and meteoritic (or mantle) sources of this highly siderophile element. The major shortcoming of this technique is its inability to distinguish meteoritic and mantle Os sources.

The first application of the Re-Os isotope system to the problem of extraterrestrial matter in sediments was by Luck and Turekian [1983] who measured the $^{187}\text{Os}/^{186}\text{Os}$ ratio in marine manganese nodules and KT boundary sediments. They found $^{187}\text{Os}/^{186}\text{Os}$ ratios in the leachable fraction of manganese

nodules to range from 6 to 8.4, indicating that dissolved Os in the oceans is derived in part from the continents and partially from extraterrestrial and/or mantle sources. Their analyses of KT boundary sediments from the Raton Basin, Colorado, and Stevns Klint, Denmark, showed $^{187}\text{Os}/^{186}\text{Os}$ ratios of 1.3 and 1.6, respectively. These values clearly rule out continental or seawater sources for the Os and are reconcilable only with an extraterrestrial or mantle source. Unfortunately, these results cannot resolve the impact versus volcanism controversy, and in fact, they illustrate that the problem cannot be explained by a simple uniform fallout layer produced by either hypothesized source. The $^{187}\text{Os}/^{186}\text{Os}$ ratios at both sites are significantly different from each other and significantly more radiogenic than expected from either a mantle or chondritic source. These $^{187}\text{Os}/^{186}\text{Os}$ ratios appear to require a small (but significant) continental contribution either from an impact target, or by precipitation, or some other plausible mechanism. This is clearly an area which deserves further research.

Recently, Esser and Turekian [1988] have applied Re-Os isotopic systematics to the background accretion rate of extraterrestrial matter. They measured the $^{187}\text{Os}/^{186}\text{Os}$ ratios in coexisting marine manganese nodules and sediments (of known accumulation rates), as well as in the leachable fraction of the sediment. These data were modeled in terms of the concentrations of three components: hydrogenous (seawater), extraterrestrial particulates, and an aeolian continental component. The continental component was modeled on an $^{187}\text{Os}/^{186}\text{Os}$ ratio of ~ 13 determined in a sample of glacial till. The hydrogenous $^{187}\text{Os}/^{186}\text{Os}$ ratio was ~ 8.5 in both the nodules and the leachable fraction of the sediment. The bulk sediment, which presumably contained extraterrestrial particulates, had a $^{187}\text{Os}/^{186}\text{Os}$ ratio of 6.5. Esser and Turekian [1988] calculated that between 62 and 70% of the Os in the bulk sediment was extraterrestrial and estimated that the extraterrestrial accretion rate was between 49 and 56 Gg a^{-1} . This is somewhat lower than the 78 Gg a^{-1} accretion rate estimated by Kyte and Wasson [1986a] but not inconsistent with their estimate, since the Esser and Turekian [1988] samples only integrated ~ 0.1 Ma of section as opposed to the ~ 30 Ma section of Kyte and Wasson [1986a] which should include a significant fraction of Ir from $\sim 10^{17}$ g objects.

The main advantage of $^{187}\text{Os}/^{186}\text{Os}$ ratios over Ir concentrations as an estimate of the extraterrestrial content of sediments is that the isotopic systematics can resolve the potential ambiguities of the fraction of continental Os (or Ir). However, this technique presently suffers from a paucity of data on the isotopic composition of continental crust which is an important factor in modeling the components in the sediment. It is also unlikely that in the near future, $^{187}\text{Os}/^{186}\text{Os}$ ratios will be determined as readily as Ir concentrations. Thus although Re-Os isotopic systematics will be valuable for application to specific problems, Ir concentrations will remain the most useful measure of extraterrestrial matter in sediments for the foreseeable future.

CONCLUDING REMARKS

The study of cosmic spherules has led to a reasonably good understanding of the types of sediment produced by the ablation of interplanetary dust. However, deposits produced by impact

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