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Extraterrestrial dust as a source of bioavailable Fe for the ocean productivity

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Abstract

Bioavailable Fe is an essential nutrient for phytoplankton that allows organisms to flourish and drawdown atmospheric CO_2 affecting global climatic condition. In marine locales remote from the continents extraterrestrial-dust provides an important source of Fe and thus moderates primary productivity. Here we provide constraints on partitioning of extraterrestrial Fe between seawater and sediments from observations of dissolution and alteration cosmic spherules recovered from the deepsea sediments and Antarctica. Of the ~3,000-6,000t/a extraterrestrial dust that reaches Earth surface, ~2–5% material survives in marine sediments whilst the remainder is liberated into seawater. Both processes contributes ~$(3\text{-}10)\times10^{-8}$molFem^{-2}yr^{-1}. Also, Fe contribution due to evaporation of survived particle is estimated to be ~10% of Fe contribution to meteoric smoke. Changes in extraterrestrial-dust flux vary not only the amount of Fe by up to three orders of magnitude, but also the partitioning of Fe between surface and abyssal waters depending on entry velocity and evaporation.

Keywords: Iron, Extraterrestrial, Micrometeorites

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1. Introduction

Iron (Fe) is the fourth most abundant element in the Earth's crust; nevertheless, it's solubility and accessibility is an essential criterion for bioavailability in isolated regions of the ocean (Johnson, 2001; Shaked et al., 2005; Norman et al., 2014). Bioavailable Fe is an essential micronutrient for the growth and survival of phytoplankton and thus impacts the drawdown of CO_2 by the oceans. It is primarily delivered by aeolian dust from continental interiors to oceans, however, it's failure to reach isolated High Nutrient Low Chlorophyll (HNLC) areas, such as Southern Ocean regions can impact the marine ecosystem and productivity (Jickells et al., 2005; Mahowald et al., 2005). Extraterrestrial dust may have a vital role in rejuvenating the biogeochemistry of the ocean for those areas where the Fe supply from aeolian dust or upwelling is scarce and is not sufficient for active primary productivity (Johnson, 2001; Reiners and Turchyn, 2018).

The heating of extraterrestrial dust during its atmospheric entry causes Fe to be added to the oceans in two distinct forms: (1) surviving particles, known as micrometeorites (MMs), that reach the Earth's surface (Genge et al., 2008), and (2) meteoritic smoke particles (MSPs) formed by recondensation of evaporated dust in the atmosphere (e.g. Hunten et al., 1980; Lanci et al., 2012). The nature of surviving particles varies considerably with particle size, entry angle and entry velocity that determines their degree of heating (Love and Brownlee, 1993). Those particles with lower entry angle into the atmosphere, however, can survive as partially melted scoriaceous MMs, or at the lowest angles survive without melting (Love and Brownlee, 1991; Rudraswami et al., 2016, 2018). With increasing size the proportion of cosmic spherules in surviving particles increases, whilst unmelted particles are most abundant at sizes of <50 µm (Genge et al., 2008). The precursor material prior to atmospheric entry, which is related to the

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source parent body, also influences the nature of the surviving MMs, for example, scoriaceous MMs form by partial melting of hydrated fine-grained matrix similar to that of carbonaceous chondrites and derived from C-type asteroids (Taylor et al., 2012; Genge et al., 2017). Extraterrestrial dust entering the atmosphere at high angles are most intensely heated and experience partial to complete evaporation (Love and Brownlee, 1991). The compositions of cosmic spherules testify to this process and show depletions in volatile and moderately volatile elements relative to chondrites. Ultimately this evaporated matter recondenses as nanometric smokes and settle to the Earth's surface (Lanci et al., 2012). Both smoke and surviving MMs are added to the oceans.

The role of extraterrestrial dust in the supply of Fe to the oceans has three significant outstanding uncertainties: (1) the average Fe content of incident dust, (2) the proportion of Fe delivered by surviving MMs compared to MSPs, and (3) the relative proportion of extraterrestrial Fe that is sequestered by seawater compared to that buried to ocean sediments. The current work presents data on the abundances, compositions and mineralogies of extraterrestrial dust recovered from deep sea sediments to address these questions. The absolute flux of extraterrestrial dust is also discussed through comparisons to other micrometeorite collections and suggests of flux of extraterrestrial Fe of up to ~10^{-6} mol Fe m^{-2} yr^{-1}.

2. Samples and Analytical Techniques

The present work has compiled micrometeorites from deep-sea sediment of central Indian Ocean and Antarctica. The deep-sea sediments collected by using surficial grab sampler (size 50×50 cm (length × breadth) from the ocean depth of ~5000 m. The second collection from Antarctica, Maitri station was undertaken by ice melting and sieving the melted water using ~50 µm mesh. The particulars of the sampling process from the deep-sea collection have been described in detail by Prasad et al. (2013) and Rudraswami et al. (2012, 2018). The micrometeorites from both these collections were mounted on epoxy for polishing to uncover the interior surface, which was carbon coated for electron microscopy studies using scanning electron microscope (SEM, JEOL JSM-IT300lV SEM with an OXFORD INCA Energy Dispersive Spectrometer detector National Institute of Oceanography, Goa) and electron Probe Micro Analyzer (EPMA, Cameca SX5). The back-scattered electron (BSE) image was obtained to classify the texture and for identification of the phases.

We have identified 5699 particles but have discarded G-type (87) and I-type (384) particles and focused our studies on S-type particles (5228). The S-type carefully chosen for the studies are as follows: scoriaceous: 220, relict-bearing: 250, porphyritic: 1710, barred: 1818, cryptocrystalline: 610, and glass: 620. All the particles are analyzed for minor and major chemical composition (Na, Mg, Si, Al, P, K, Ca, Ti, Cr, Fe, Ni, Mn) using electron microprobe to obtain bulk chemical composition which are provided in Supplementary file A. Analyses on each particle varied from ~10 to 20 spots having a beam size of ~2−5 µm with gaps between each point such that the entire particle was covered representing true bulk composition. The specifics related to the electron microscopy analyses technique is discussed elsewhere (Rudraswami et al., 2011; Rudraswami et al. 2012). These studies are focused on particle sizes that deliver the mass peak of the ET matter to Earth.

3. Results and Discussion

3.1. The type of micrometeorite and average Fe-content

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The types of MMs recovered on Earth vary significantly with particle size. At diameters of ~200 μm studies of unmelted MMs recovered from Antarctic ice reveal that the majority are dominated by materials with fine-grained hydrated precursors similar to the CI and CM chondrites, with a smaller proportion of CR materials (Genge et al., 2008; Taylor et al., 2012). Crystalline MMs present are dominated by olivine, pyroxene and glass. These particles are thought to be fragments of chondrules from chondritic parent bodies and include particles from ordinary chondrites (OCs; Genge, 2008; Prasad et al., 2015). Although unmelted particles represent a small fraction of MMs in this size range, they are representative of the flux since they survive by virtue of their low entry angles, thus cosmic spherules will have the same sources. Using the bulk compositions of different parent bodies the Fe contribution from each parental type of MM that is calculated is shown in Fig. 1. We have used various composition of chondritic Fe content which is provided in Hutchison (2004), and Johnson (2001) formula to calculate the total Fe for various chondrites to generate the Table 1. The formula is as follows: Total bioavailable input to ocean = [(Extraterrestrial Flux) × 0.9 (considering 90% ablation) × 0.7 (70% area of Earth is covered by ocean) × (weight percent of iron of various chondrites)] / [area of ocean × molar mass of Fe]. The area of global ocean considered for this study is ~3.62 × 108 km^2. We have incorporated 90% ablation for ground based collection technique to calculate the total flux before atmospheric entry. This 90% ablation is based on Taylor et al. (1998) where they have considered Love and Brownlee (1993) flux data from experiments done on Long Duration Exposure Facility (LDEF) at an orbital altitude of ~300–400 km.

Observations of large MMs (>300 μm) from the Transantarctic mountains suggest that the contribution of ordinary chondrites becomes higher (~30%) at large sizes (Cordier et al., 2011; Van Ginneken et al., 2017) will increase the average Fe content. Since particle abundance

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decreases exponentially with size, however, the increase in the average will be small. Small MMs are dominated by hydrous particles similar to carbonaceous chondrites, but contain ~40% carbon-rich, porous cometary particles (Noguchi et al., 2015). The abundance of Fe within cometary porous particles is lower than in chondrites since it is diluted by abundant carbonaceous matter (>30%). The increase in average Fe owing to large OC particles is, therefore, offset by its lower content in small cometary grains. The parent body abundances amongst Indian Ocean cosmic spherules, therefore, give a good estimate for the average Fe abundance of the ET flux prior to atmospheric entry.

3.2. Relative importance of smokes and micrometeorites

Numerical models of atmospheric entry suggest that ~90% of the Extraterrestrial dust mass flux is lost to evaporation during atmospheric entry (Love and Brownlee, 1993). MMs collected from Antarctic snow at the South Pole Water Well provide a means of evaluating the proportion of mass lost to evaporation (Taylor et al., 1998). Different researchers using various collection techniques have arrived at different flux rate. It was estimated based on courting statistics by Taylor et al. (1998) from South Pole Water Well (SPWW) that ~3000 tonnes per annum survive the heating during atmospheric entry and reach the Earth surface indicating that more than ~90% that get ablated does not make it to Earth's surface. This estimate of particles reaching the Earth surface is lower than that estimated by Yada et al. (2004) who has coupled the handpicking of micrometeorites and noble gas measurement from the residue, and has indicated a range of 11000–16000 tonnes per annum as the extraterrestrial material reaching the Earth surface. However, Yada et al. (2004) based only on handpicked particle count has shown ~5000–7000 tonnes per annum. This is much higher than those estimated by previous researchers, probably one of the reason is the smaller sieved size used by Yada et al. (2004).

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Nevertheless, the micrometeorites from deep-sea sediments are much lower than Antarctica (Brownlee et al., 1997; Prasad et al., 2013). Measurements of micro-impact craters on the LDEF satellite provide arguably the most accurate observational evidence for the total flux of Extraterrestrial dust prior to atmospheric entry at 40,000 t a^{-1} (Love and Brownlee, 1993). The total fraction of evaporated material is thus 80–90% with a most likely value of ~90% similar to that predicted by models of entry heating.

Partial evaporation must also be considered when evaluating the proportion of Fe liberated by evaporation and the formation of smokes. The composition of 5228 deepsea and Antarctica spherules analysed in the current study is shown in Fig. 2 (data in supplementary file A) based on their texture classification which suggests preferential loss of Fe compared to more refractory elements such as Al, Ca, Mg, and Si. An average Fe content of ~20 wt% is retained in Indian Ocean spherules. Since these particles are the surviving remnants of evaporation, the fractionation of Fe during entry heating can be estimated by comparison to the predicted precursor average Fe content of ~23% (carbonaceous and ordinary chondrites). This suggests that MSPs that contribute ~2 t d^{-1} (Hervig et al., 2017) are enriched in Fe by comparison to their precursor compositions by a factor of 3–7 times based on the ground based collection techniques (Taylor et al., 1998; Yada et al., 2004). Hence, the overall Fe contribution based on particle chemical analyses is estimated be ~0.2–0.5 t d^{-1} which is ~10% of MSPs contribution. Dhomse et al. (2013) studied the transport of MSPs and their deposition to the earth's surface and predicted preferential deposition occurs at mid-latitudes by a factor of ~10 compared to other locales. Nevertheless, Lanci et al. (2012) showed that nanometer size supramagnetic iron exists within polar ice indicating that the recondensed evaporated portion of incoming ET material does reach the Earth’s surface at all latitudes.

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3.3. Burial of extraterrestrial Fe in Ocean sediments

Many previous researchers related to the topic of "Fe source to Ocean by extraterrestrial input" has focused only on the total material that is hit above the Earth's surface ignoring the contribution from the micrometeorites that fall on Earth surface. The sample that is retained form the Earth surface have evaporated nearly ~10 to 80% of their original mass, besides, etching that it undergoes in the ocean contributing effectively to marine biogeochemistry. Most of the preserved sample from the ocean are etched compared to inside portion as represented in Figure 1. Ignoring the size of the spherule, the percent of spherule etched out in different types of spherules varies given the age (~0–50,000 years) of the sediment from which the sample is collected (Prasad et al., 2013). The etching is commonly observed in barred spherules due to hydration. The more the time spherule spent in seawater, the more it gets etched out. The sample once gets settled in the sediments the chances of etching diminishes. All the spherules show varying levels of etching considering their large range in terrestrial ages, except, the barred spherules that show maximum etching among all textural types. The etching phenomena is commonly seen in any collection, whether it is Antarctica or deep-sea sediments, while it is dominant in later due to large residence time and harsh condition of ocean. The glass spherules have shown minimum alteration compared to its counterpart due low porosity, and highly melted during atmospheric entry. Papanastassiou et al. (1983) observed no chemical alteration in measured $^{87}Sr/^{86}Sr$ ratios and the Sr concentration in the analyzed deep-sea spherules have typical chondritic range suggesting that they have not exchanged Sr with seawater. The Rb depletion in spherules is related to volatilization during atmospheric entry and has not related to weathering in seawater. They concluded that the $^{87}8r/^{86}Sr$ in deep-sea spherules is consistent with origin from chondritic composition meteoroids and appears to exclude most other possible terrestrial sources. Also extensive study of nearly ~300 spherules

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done by Bate et al. (1986) drawn conclusion that the average Mg/Si, Al/Si and Ca/Si ratios of the deep-sea spherule has not changed with time scale in spite of etching.

The abundance of deep sea spherules collected in the Indian Ocean provides a direct measurement of the proportion of ET Fe buried within ocean sediments. The deficit between the flux of MMs retained in sediments and the flux at the top of the atmosphere relates to the mass lost to evaporation, plus the mass of MMs dissolved into seawater. The mass lost to evaporation was suggested above to be ~90%. Values of the spherule flux of ~160 t a^{-1} from the Indian Ocean collection (Prasad et al., 2013), therefore, suggest the mass of Fe released by dissolution into seawater is ~5–10% of the total incoming Fe flux (>90% of MMs destroyed by dissolution). The degree of dissolution of MMs through alteration in seawater can be independently confirmed by the relative abundance of I-type cosmic spherules. These particles form by atmospheric entry and oxidation of metal grains to magnetite (Fe_3O_4) and wustite (FeO). Both these minerals are highly resistant to aqueous alteration compared with silicates. The relative abundance of I-types to S-type (silicate) spherules, therefore, gives a measure of proportion of S-types destroyed by dissolution. The abundance of I-types in the Indian Ocean collection is ~5% whilst that in the South Pole Water Well is ~1% suggesting the loss of ~80% of S-type spherules by dissolution in the ocean, broadly consistent with the fraction predicted from the over-all spherule abundance.

Although ~80–90% of deep sea spherules can be inferred to have been destroyed by dissolution, surviving particles also exhibit evidence for significant etching which varies from 20 to 90% by volume (Fig. 3). S-type cosmic spherules often show the removal of interstitial glass between crystals of olivine with etching concentrated in external rims (Fig. 3). Glass in these particles is Fe-rich in comparison to olivine, which is usually Mg-rich, consequently

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etching causes loss of Fe. The rough estimate based on Taylor et al. (1998) or Yada et al. (2004) flux calculation indicate that ~3,000–16,000 t a^{-1} of ET material get dissolved bring ~25–30% of Fe from total flux depending on different types of precursors. This will add at least ~(3–10)×10^{-8} mol Fe m^{-2} yr^{-1}. However, particles etching of ~20–90% in seawater contribute few percent to the above value.

4. Implications

Data from the Indian Ocean collection of deep sea spherules suggests that slightly more than 90% of Fe within extraterrestrial dust incident on the atmosphere is lost from particles during evaporation and delivered to the ocean as recondensed MSPs. Amongst the 10% of extraterrestrial dust that survives atmospheric entry >90% of the initial flux of Fe is liberated by dissolution of MMs in seawater, with 2–5% buried within deep sea sediments and thus not bioavailable. Given a measured flux of 40,000 t a^{-1} this suggests an average 3×10^{-7} mol Fe m^{-2} yr^{-1} of Fe delivered to the oceans. Some simulations of the flux of dust delivered to the Earth, based on its orbital evolution, however, suggest a maximum of ~10^5 t a^{-1} (Nesvorný et al., 2010) and approximately double the Fe flux. Estimates of the flux of terrestrial dust to the Southern Ocean give a total mass of 30×10^{-6} mol Fe m^{-2} yr^{-1} (Lancelot et al., 2009), however, silicic volcanic dust and wind-blown clastic grains dominated by quartz are both Fe-poor. Furthermore, Fe in such terrestrial dust particles is present as Fe oxides that are relatively resistant to dissolution in seawater. The soluble fraction is likely to be <1%. The ET Fe flux is, therefore, likely to be similar to that from terrestrial sources. In the Southern Ocean concentration of MSPs is also increased by a factor of 10 and is likely exceed terrestrial sources and dominate primary production of phytoplankton (Dhomse et al., 2013). Elsewhere MSP are

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less abundant and Fe from surviving particles represent a larger fraction (~20%) of extraterrestrial Fe.

The nature of ET inputs of Fe to the oceans, however, are also crucially important. Meteoritic smokes represent the largest input to the oceans and are dominated by nanometric dust (Lanci et al., 2012), which are likely to be readily soluble owing to their high surface area to mass ratio. Smokes are added directly to the ocean surface and rapidly release their Fe within the photic zone. This Fe is, therefore, immediately available to phytoplankton. At low sedimentation rates of a few microns per year on parts of the abyssal plain of settled particles are exposed to seawater where Fe get leached out by dissolution of MMs at abyssal depths. The Fe derived by dissolution of MMs, therefore, is not immediately available to phytoplankton but is stored at depth until upwelling currents bring it to the surface. In areas distal to the continents, such as isolated islands and seamounts, and the intertropical convergence zone, extraterrestrial Fe may impact primary production.

Currently the importance of extraterrestrial Fe in primary production is likely to be localised within those areas with the lowest influx of terrestrial dust. The ET flux of dust, however, has been elevated at times in the Earth's past such as in the Ordovician (Schmitz et al., 2019) and at the end of the Eocene (Meier et al., 2016), with increases in flux up to 2–3 orders of magnitude. During these periods extraterrestrial Fe delivered to the oceans may have dominated primary production more widely and thus affected global CO_2 budget. This mechanism, together with atmospheric dust load, has already been suggested as an origin for the end Ordovician glaciation (Schmitz et al., 2019). Evidence also exists for periodic changes in the ET flux the eccentricity variations in the Earth's orbit. Periods of higher entry velocity will enhance evaporation of dust and delivery of Fe to the photic zone of the ocean, whilst lower entry velocity will increase Fe delivered to abyssal waters that replenish the surface by subsequent vertical mixing. The velocity distribution of extraterrestrial dust, therefore, will influence the consequences of the delivery of extraterrestrial Fe to Earth.

5. Conclusion

Data from the abundance, composition and textures of deep sea spherules allow the first assessment partitioning of extraterrestrial Fe between MSPs, surviving micrometeorites, marine sediments and ocean water. The abundance of extraterrestrial Fe liberated by evaporation is suggested to be enhanced owing to the effects of partial evaporation and forms part of the component delivered to the oceans as MSPs. This material dominates the flux of extraterrestrial Fe delivered to the oceans and is dissolved in the photic zone of the oceans where it directly impacts primary productivity. Surviving MMs in contrast undergo 80–90% dissolution in the oceans and their Fe is delivered to abyssal depths. Upwelling of seawater from depth allows this extraterrestrial Fe to contribute to primary productivity. At present extraterrestrial Fe is important in areas where the supply of terrestrial dust is low, such as in the Southern Oceans, however, during periods in which ET flux is enhanced it is likely to have a wider-spread contribution to productivity and thus CO_2 drawdown by the oceans affecting global climate.

Data availability. The supporting data (supplementary A) related to this article is available at Dryad Dataset (https://datadryad.org/stash/dataset/doi:10.5061/dryad.zcrjdfn7t).

Author contributions. NGR is involved in collecting micrometeroites, project funding, formulating the research objective. MP is involved in compilation of data and preparation of the manuscript. DF is involved in collection and analysis of data. MJG is involved in preparation of the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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FIGURE CAPTIONS

Figure 1.

Evaluating the contribution of iron from various extraterrestrial chondritic precursors having different Fe content and flux rate estimated by various researchers adopting different methodologies (Table 1). The estimate are done by assuming 90% ablation for the calculated flux by various researchers. It can be seen that Yada et al. (2004) flux shown huge contribution due top more flux reported by them in their collection, while on the other hand the deep-sea collection has lower estimate as a large fraction get etched out in seawater. The bulk Fe content of various chondrites is given by Hutchison (2004).

Figure 2.

The plot of major element Fe versus Si and Mg for different type of cosmic spherules, namely, scoriaceous (220), relict-bearing (250), porphyritic (1710), barred (1818), cryptocrystalline (610), and glass (620), respectively.

Figure 3.

Back-scattered electron images of the cosmic spherules that have various level of etching collected from deep sea sediments of Indian Ocean.

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Fig. 1

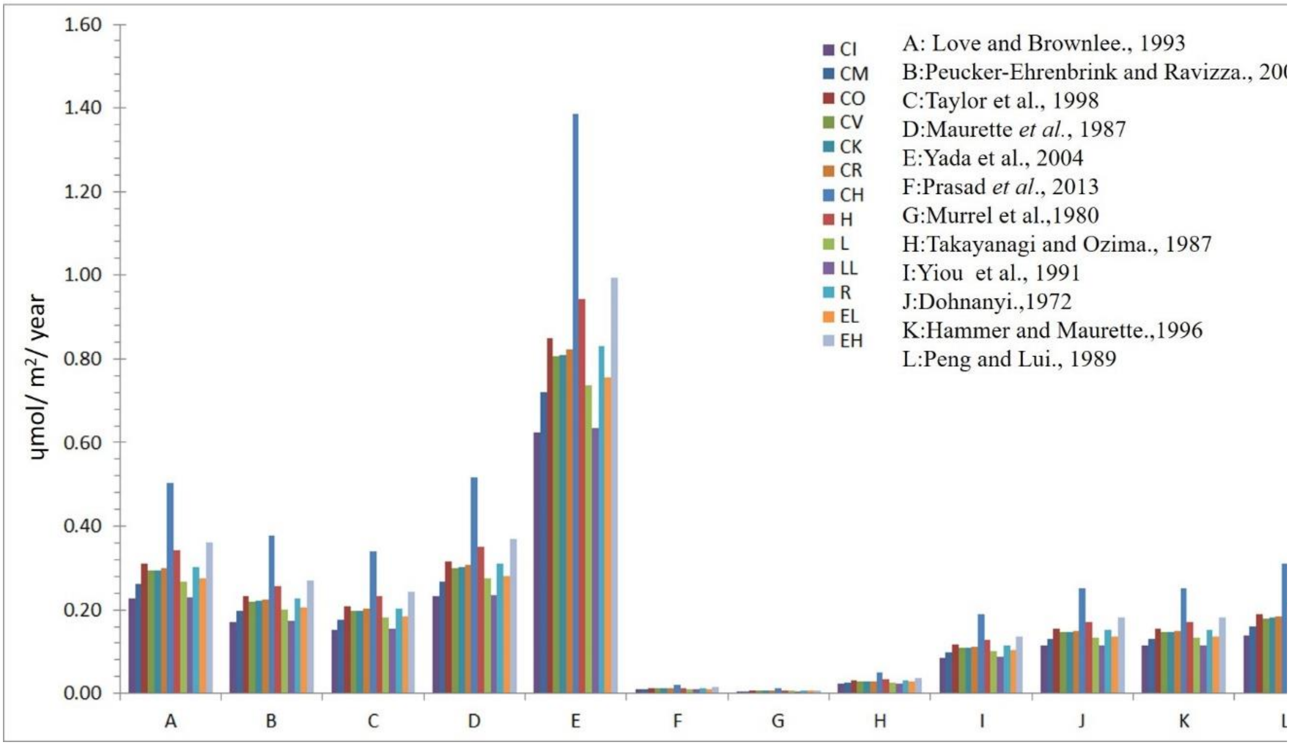

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Fig. 2

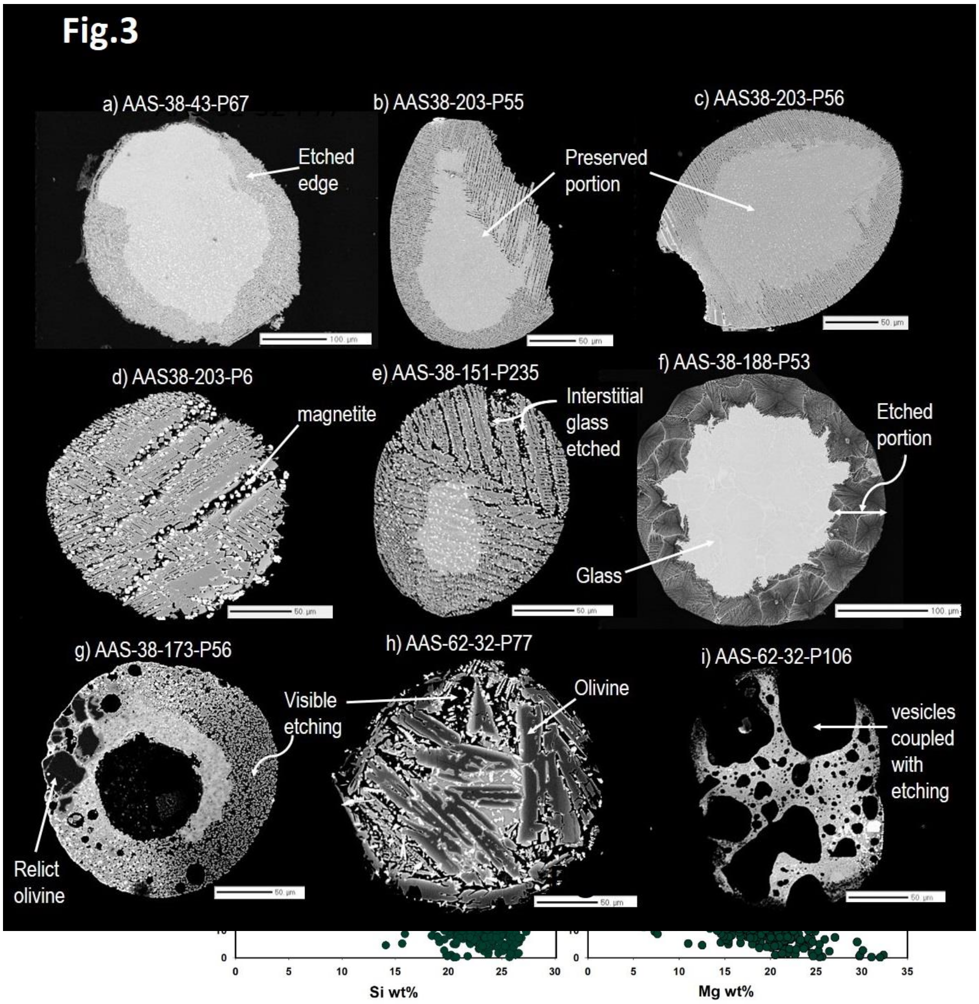

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Table 1. The total Fe input to ocean based on different type of chondrites has been calculated using data from Hutchison (2004) and formula based on Johnson (2001). The ablation of MMs during entry is considered to be ~90% based on Taylor et al. (1998) and Love and Bronwlee (1993). The values in the table are in umol Fe m^{-2} yr^{-1}.

	Total Fe for CI	**Total Fe for CM**	**Total Fe for CO**	**Total Fe for CV**	**Total Fe for CK**	**Total Fe for CR**	**Total Fe for CH**	**Total Fe for H**	**Total Fe for L**	**Total Fe for LL**	**Total Fe for R**	**Total Fe for EL**	**Total Fe for EH**	
A	0.23	0.26	0.31	0.29	0.29	0.30	0.50	0.34	0.27	0.23	0.30	0.27	0.36	Love and Brownlee., 1993
B	0.17	0.20	0.23	0.22	0.22	0.22	0.38	0.26	0.20	0.17	0.23	0.21	0.27	Peucker-Ehrenbrink and Ravizza, 2000
C	0.15	0.18	0.21	0.20	0.20	0.20	0.34	0.23	0.18	0.16	0.20	0.19	0.24	Taylor et al., 1998
D	0.23	0.27	0.32	0.30	0.30	0.31	0.52	0.35	0.27	0.24	0.31	0.28	0.37	Maurette *et al.*, 1987
E	0.62	0.72	0.85	0.81	0.81	0.82	1.38	0.94	0.74	0.63	0.83	0.75	0.99	Yada et al. 2004
F	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	Prasad *et al*. 2013
G	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	Murrel et al.1980
H	0.02	0.03	0.03	0.03	0.03	0.03	0.05	0.03	0.03	0.02	0.03	0.03	0.04	Takayanagi and Ozima, 1987
I	0.09	0.10	0.12	0.11	0.11	0.11	0.19	0.13	0.10	0.09	0.11	0.10	0.14	Yiou et al. 1991
J	0.11	0.13	0.15	0.15	0.15	0.15	0.25	0.17	0.13	0.12	0.15	0.14	0.18	Dohnanyi.1972
K	0.11	0.13	0.15	0.15	0.15	0.15	0.25	0.17	0.13	0.12	0.15	0.14	0.18	Hammer and Maurette, 1996
L	0.14	0.16	0.19	0.18	0.18	0.18	0.31	0.21	0.16	0.14	0.19	0.17	0.22	Peng and Lui 1989