

Overview:

This manuscript presents results from the Indian Ocean on a study estimating the potential of extraterrestrial dust as a source of bioavailable iron to the open ocean for fuelling ocean productivity. While the study is interesting it currently has a number of fatal flaws in its approach that make it unpublishable in its present form as detailed below. Oddly in reading the paper it is not clear that they even needed their data on micrometeorites from the Indian Ocean as it is not used directly in any of the calculations on the bioavailable iron made in the paper as all the terms used are derived from other works. The only term that this data could be used for is the iron percent composition of the micrometeorites and in this work that is derived from (Prasad et al., 2013) and gives the lowest flux values, and this is not commented on in the paper, so it just leaves the reader quite perplexed about how all this is meant to come together in a coherent fashion.

General Comments:

Not all iron is bioavailable – A frequent error in this paper is to use the term bioavailable but without any context, a definition of some sort needs to be provided, there are a number of studies that have tried to provide some sort of basis for this with respect to bioavailable iron (Croot and Heller, 2012; Eldridge et al., 2004; Fan et al., 2006; Hudson et al., 1992; Hudson and Morel, 1990, 1993; Ito, 2013; Lam et al., 2006; Raiswell et al., 2008; Suzuki et al., 1995). In the present work, many times all iron is considered bioavailable and the issue of the chemistry is not included in any analysis and so the paper grossly overestimates the impact of the extraterrestrial Fe to the ocean.

Is iron ablated from extraterrestrial dust bioavailable? – The question of whether the iron that was removed from micrometeorites (MMs) as they enter the atmosphere is bioavailable or not, was not really discussed in this work. Saunders and Plane (2006) suggested that the iron in meteoritic smoke particles (MSPs) was comprised of the minerals hematite (Fe_2O_3), goethite (FeOOH) and fayalite (Fe_2SiO_4), all of which are poorly bioavailable in seawater, even as colloids or nanoparticles (Nodwell and Price, 2001; Wells, 1989, 1999). Though other studies have suggested that meteoritic iron may or may not be found in the dissolved phase in the lower stratosphere depending on the sulfuric acid concentration (Murphy et al., 2014). That study also goes on to suggest that transport through the troposphere would lead to these particles becoming partially solid as they are oxidized or neutralized (Froyd et al., 2009). Work by Lanci et al. (2007) where the meteoric smoke concentration in the Vostok ice core was determined assumed that the signal they were observing was from iron in nano magnetite particles – which are also not readily bioavailable, though mixotrophic organisms may be able to utilize it (Nodwell and Price, 2001).

Iron dissolution in the water column: A critical weakness of this paper is that it assumes that the iron missing from the sediments in terms of the MM (or MSP) mass balance has been dissolved. The numbers that the authors calculate don't consider that this iron is deposited to the sediment, but it is distinct from the unmelted or partially melted MM particles and thus not considered in their mass balance, this appears to be a massive oversight on the part of the authors. The chemistry of these particles argues very much against them being highly soluble in seawater (as explained above) and so the most likely outcome is that this material is in the sediment but it indistinguishable, to the most part, from continental dust. The MMs that are in the sediments are clearly not very soluble so if they are model particles for the dissolution in the water column then this would be more than likely lower than 0.1% in terms of the fractional solubility (Baker and Croot, 2010).

Numeracy and errors – Throughout this manuscript it is often very hard to follow how the calculations were made and if any error bars were considered, this is particularly important when it

comes to how the dissolution calculations are made as many of the numbers seem to come out of thin air and no evidence for them is presented in the current manuscript. This may be an issue with the language used to describe how the calculation was made, and thus could be easily improved, at present this aspect of the paper is also a serious weakness.

Specific Comments:

P2 line 13: Fe is an essential nutrient, the adjective bioavailable confuses things in this opening sentence as it tries to make a distinction that is not needed at this point. See the general comment above regarding the use of the term bioavailable iron throughout this work.

P2 line 15: continents, extraterrestrial dust

P2 line 20: Liberated does not seem a good way to describe it, as it is likely precipitated in the water column and then removed to the sediments. However now it is not detectable as a distinct micro-meteorite.

P2 line 21: Of iron, the amount that is bioavailable is not known.

P2 line 21: mol Fe m⁻² yr⁻¹ (space this out, don't run it together)

P2 line 21: This is a very small fraction of the total iron deposition from dust to the oceans (Jickells et al., 2005) as even in the regions furthest from the continental margins (e.g. South Pacific) the deposition rate is considered to be 0.05-0.1 g m⁻² y⁻¹ (Pavia et al., 2020) and considering an Fe content of 3.5% (Rudnick and Gao, 2003; Taylor and McLennan, 1985; Taylor and McLennan, 1995) that converts to 31.3 – 62.7 μmol Fe m⁻² y⁻¹. This simple calculation then indicates that only 0.05-0.32 % of the total flux is potentially sourced from extraterrestrial dust – is this really significant?

P2 line 23: What time scale are the changes referred to here over? The 3 order of magnitude change is the extraterrestrial dust flux itself? If so over what spatial scales? This final sentence seems very vague and imprecise and should be refocused.

P3 line 34: What is meant by accessibility in this sentence? Reactivity or lability seems to be more of a key parameter with respect to bioavailability (Croot and Heller, 2012; Hudson, 1989; Hudson and Morel, 1993).

P3 line 35: As for P2 line 13, bioavailable iron is not a micronutrient, iron is a micronutrient.

P3 line 37: There are other mechanisms as well (Tagliabue et al., 2010; Tagliabue and Resing, 2016; Tagliabue et al., 2014a; Tagliabue et al., 2014b) and they should be summarized here.

P4 lines 60-64: Nothing in this section addresses the issue of iron bioavailability. See the general comment above.

P4 line 69: Sequestered would mean that it was retained in the seawater – no evidence has been provided for that nor a mechanism provided, strongly suggest this be rephrased. Most of the continental dust makes its ultimately into the sediments so why would the iron from MMs and MSPs be any different?

P4 line 73: This is at least getting closer to the terrestrial aerosol flux over the oceans but it needs a citation as it appears to come out of nowhere.

P5 line 90-91: It would be useful for this audience if you explained what G-type, I-type and S-type particles are in this context and/or gave a reference to this. Presumably the authors are using the classification from Genge et al. (2008). Does this analysis include any of the reportedly unmelted micrometeorites found at the same site? (Genge and Van Ginneken, 2018; Prasad et al., 2017; Prasad et al., 2018).

P5 line 91: What does careful mean in this sense? What criteria were used for choosing the particles?

P5 line 99: What particle sizes are these? Can you at least give the reader some values? As they seem to be very large (10's of microns) if you can have multiple 2-5 μm spots analyzed on the particle.

P6 Line 111: See the earlier comment regarding the presence of unmelted micrometeorites (Genge and Van Ginneken, 2018; Prasad et al., 2017; Prasad et al., 2018), what is the proportion of unmelted micrometeorites in these samples?

P6 line 118: This calculation assumes that all of the iron is bioavailable, which is something that is very unlikely, the true bioavailable iron would be on the order of 1% or less than this if the solid phase is magnetite or goethite etc. See the general comment above on this. I strongly suggest you remove the word bioavailable from this sentence.

P6 Line 119: What was the weight percent of iron used for this calculation? It needs to be specified here even if it is just a range of values.

P6 line 121: 10^8 km^2

P6 line 122: This is a much higher number than indicated in earlier works by one of the authors of this work (Rudraswami et al., 2015), where 65% of Antarctic micrometeorites underwent less than 20% ablation. Can the authors provide some context as to why they went for the 90% value rather than the lower values seen in other studies?

P7 line 145: counting

P7 line 141: So where does Rudraswami et al. (2015) fit into this scheme?

P8 line 155: lower in terms of what? Iron content, altitude, latitude? This sentence needs to be better constructed.

P8 line 158: Better to be consistent and use t yr^{-1} than t a^{-1} unless the a means something else in the context?

P9 line 185: So not 90% as was used in the calculations described above?

P9 line 188-206: This whole section on 'etching' could be removed as it does not add anything to the paper. Simply put any post depositional change to the micrometeorite in the sediment is not going to make much of a difference to sediment fluxes.

P10 line 212: This is a critical value but the justification for it seems to be very poor and other evidence is contradictory.

P10 line 212: Same comment as for line 158.

P10 line 212: Does this value of 160 t yr⁻¹ refer to the Indian Ocean or the whole globe? In the cited paper (Prasad et al., 2013) it is apparently the whole global but that is not reported in the present manuscript.

P10 line 214: This is hard to follow, 90% is lost to the atmosphere directly, but still will arrive to the sea at some point, the 5-10% dissolution is based on the total flux. This is an extremely high solubility for an iron phase and it is hard to match this up with the phases that have been suggested in the literature. It also begs the question of what the error bars are on the estimate of the spherule flux and what this is on a m⁻² basis (Prasad et al., 2013). Importantly how was the 5-10% number arrived at in the end as this is not at all obvious and difficult to see how when on line 158 the flux in is 40,000 t yr⁻¹ and 70% of this is over the ocean and 90% of that is lost then that is 4,000 t yr⁻¹ arriving at the ocean without modification and if only 160 t yr⁻¹ are deposited as MM in the sediment that leaves a deficit of 3840 t yr⁻¹ that is apparently dissolved using this line of argument, which is approx. 10% of the total flux but only because of the 90% estimate for the atmosphere, so there isn't any independent estimate here. It should be noted that the other 36,000 t yr⁻¹ converted to other iron forms in the atmosphere is presumably also deposited to the earth's surface.

P10 line 219-223: This seems a very simplistic way to gauge the dissolution of S-type spherules, and there is a logic error here also – the first sentence talks about the ratio of I to S but the calculation is done on the abundance of I types, not the ratio, so it assumes then that there is only I and S types. It also ignores the evidence that there is a high abundance of I types in the sediments (based on the data provided here) so this unfortunately appears to be a flawed way to approach this. I would suggest removing this section as it currently is extremely weak and detracts from the rest of the paper.

P11 line 238: This statement is not correct, as the 90% value comes not from this work but from a cited reference (Taylor et al., 1998), more importantly no evidence is shown in this work for any particles that are recondensed MSPs. This sentence then needs to be altered to reflect what was actually presented here.

P11 line 241: This is an absolutely incredible number you are suggesting, that 90% of the iron that enters the ocean as MMs is dissolved. It just is not supported by the chemistry and instead looks very much like it is just not accounted for in the sediments and that the dissolution is massively overestimated.

P11 line 242: Is this 2-5% buried of the total flux or the flux that was not ablated in the atmosphere?

P11 line 243: as for previous comments, better to use yr⁻¹ than a⁻¹. In this sentence you use both so it is could be easily confused.

P11 line 243: How was the iron value calculated here as this number does not match the inputs given above?

P11 line 247: Is this the dust flux to the whole Southern Ocean or to the Indian Ocean sector of the Southern Ocean? It makes a difference as there are other sources to the different sectors of the Southern Ocean (Bullard et al., 2016; Gassó and Stein, 2007; Johnson et al., 2011; Johnson et al., 2010; Li et al., 2008; Neff and Bertler, 2015; Tatlhego et al., 2020).

P11 line 248: This sentence should be rephrased as you provide an Fe estimate and then discuss how iron poor the dust is, this would only be relevant if only the dust flux was provided.

P11 line 250: Please provide a citation for this figure.

P11 line 250: In this context what is the fractional solubility for the MM and MSP derived iron? See the general comment on this above.

P11 line 252: So the bulk of aerosol deposition here is from MSPs? What evidence is there for this in the dust aerosols over the Southern Ocean? It seems based on this study it should be full of iron from MSPs, though this does not seem to have been observed. The cited reference (Dhomse et al., 2013) uses data from Saunders et al. (2012) which suggested that all of the Fe was in the form of highly soluble iron sulfates in the stratosphere, however other studies suggest that the iron would be in less soluble forms once it has passed through the troposphere (see general comment above), as it must to get to the ocean surface.

P11 line 252: It needs to be remembered that the MSP and MM flux is part of the overall dust flux and not something that can be thought of as extra to the measured dust fluxes.

P12 line 259: A small particle size does give a high surface area to volume ratio and this will increase solubility but the key aspect here is the chemical form of the iron in the particle and most of the data that is published indicates that it would be very insoluble. It also needs to be noted that the particles in question here are large relative to where these solubility effects would become important, as the particles here are much larger than colloidal or nanoparticles given they have sizes of 10's of microns as MMs.

P12 line 260: Do you have any data on the size range of MSPs that would support this suggestion? Presumably they are rained out the same as terrestrial dust and so could be as either wet or dry deposition.

P12 line 261-263: It is not clear what this sentence is trying to suggest? Is it that MMs would sink very slowly in the water column despite their size and density?

P12 line 265-267: This is analogous to the arguments for hydrothermal iron (Tagliabue et al., 2010; Tagliabue and Resing, 2016) which is a significantly larger flux to the ocean.

P20-21,23: It should be explained what each of the chondrites types are as there isn't anything like that in the original paper by Johnson (2001) and it makes for a very confusing table and figure. Given also all the data that is provided in the supplementary information where does the current dataset plot here?

P23: Given that the data in this study seems to have been derived from the same work as (Prasad et al., 2013) and this gives the lowest estimates for the iron input to the ocean, some explanation is required as this paper seems to ignore that particular result and use the higher ranges provided by other works.

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