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Discussions

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Interactive comment on "Mechanisms of dissolved and labile particulate iron supply to shelf waters and phytoplankton blooms off South Georgia, Southern Ocean" *by* Christian Schlosser et al.

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We thank the reviewer for their comments, but think that important findings of this paper were misunderstood or overlooked. For that reason we just conducted minor changes. This paper continuous the recently published paper from Schmidt et al. [2016] and highlights the strength of the Fe supply from the South Georgia shelf by applying three independent data sets collected in 2011 and 2013 on the shelf and shelf edge of South Georgia. The supply of dissolved Fe form shelf sediments, glacial outflow and zoo-plankton activity on the shallow shelf is discussed in section 3.1, 3.2 and 3.3. Section 3.4 discusses the supply of DFe in the bloom region north of the island, ca. 1,250 km

downstream the South Georgia source region. This is stated in the paragraph, line 429ff. Oceanographic settings at our study site on the South Georgia shelf (source region) are entirely different to open ocean sites downstream the island (Fe budget in the bloom region). Vertical diffusive and advective mixing are much stronger on the shelf than off-shore. For instance, internal waves that cross the 200 m deep shelf create strong turbulences that facilitate the diffusive and advective term of vertical mixing (upwelling, references are listed below). Father off shore in the bloom region, the vertical Fe supply is limited because of a deep ferrocline, reduced diffusive mixing and negative Eckman transport (downwelling [Tagliabue et al., 2014]). Recent publications illustrate that DFe supplied from continental or island arc shelf sediments drive offshore primary productivity [Graham et al., 2015]. The intriguing question remains, how DFe from the source region (continental shelf and island arc) is transported downstream in the region with elevated photosynthetic activity (bloom region). This shelf or island effect can be found on numerous islands in the Southern Ocean [Graham et al., 2015]. Although we mainly rely on literature values for the bloom region we estimated that the Fe demand in the bloom region is higher than the actual Fe supply (section 3.4). We suggested that the missing DFe quote must be delivered from biogenic Fe particles that were created under high Fe conditions on the South Georgia shelf and Fe was released downstream the island by internal recycling in the mixing layer. This process has been discussed in detail line 516ff. Please find the comments of the reviewer, and our reply to it underneath (Answer:.. ..).

Specific comments: Methods: -Why were sediment cores collected on the southern shelf and not the northern shelf? Given that the phytoplankton bloom appears to original on the northern shelf, and all water column and particle samples were collected on the northern shelf, this seems like a strange decision. If this was outside of the authors' control, then this needs to at least be discussed when sediments are compared to the water column, since one would expect the stronger bloom on the northside to affect the benthic processing of Fe and Mn, so sediments on the southern shelf may not be representative of those on the northern shelf. Answer: We agree that sediment

cores from the productive northern side of the island would be better. However, JC055 was a benthic cruise, hunting gas hydrates, and thus sampling locations were outside our control. So, you can't get always what you want! We agree that an elevated organic content in marine sediments would alter substantially benthic redox processes and usually increases the content of Fe(II) and Mn(II) in pore waters and thus facilitates the efflux of sedimentary Fe. This may mean that the bentic Fe supply on the northern side is probably higher and the efflux on the southern side (0.1 – 44 μmol m-2 d-1) underestimates the northern shelf efflux. However, this has no effect on our vertical Fe supply calculations in surface waters on the northern side (see supplementary text). To clarify this issue we added a sentence in the section 2.1 (line 117ff) and section 3.2.1 (line 291ff).

-Fecal pellet data are first discussed in section 3.1.3 (for data in Table 4) but without introducing the methodology for fecal pellet sampling and analysis. They are discussed again in section 3.2.3, where a reference is to the Schmidt et al papers, but this should be moved up. Answer: To clarify we added "as described by Schmidt et al. [2016]" (line 254ff) and added "and fecal pellets" to the header of section 2.2 (line 126).

Section 3.1.1 -The discussion of particulate fractions in this section is confusing. LPun is derived from acidified unfiltered seawater and defined in equation 1, but then is compared to P as if they should be the same, differing only in their sampling methods (acidified unfiltered seawater vs SAPS sampleâËŸAËĞ Tlines 157-158). However, P is defined in section 2.2 as the sum of leachable (LP) and refractory (RP) fractions from the SAPS samples, so one wouldn't expect LPun and P to be the same. I initially read this section thinking that they were comparing LPun (from acidified unfiltered seawater) and LP (from acetic acid leach of SAPS samples), which is the more direct comparison if they want to isolate sampling differences (but still not perfect since acidification to pH 1.7 with HNO3 is still not the same as a 25% acetic acid leach, but at least more comparable). This section needs to be clarified. It seems that they have two points in this section: 1) that pFe and pAl have a refractory component, since LP(un)/P < 100%,

and 2) LP(un) scales with P, so they want to justify using LP(un) for P. For the first point, it seems that a wholly SAPS-derived assessment of LP/(LP+RP) would be the better parameter to present, because then there is no confusion of mixing sampling systems, pore sizes, and leach types. This is done in section 3.2.2 (lines 320-340), which would make more sense in this section Answer: We disagree with the reviewer. We observed that Iron, Mn, and Al in the LPUn and P fraction show a linear relationship (see Fig. 3 and Table 1and 2) and that highest PFe concentration coincide with slightly lower but comparable LPUnFe concentrations in the water column (Fig. 2). This is not surprising (see details below), but we wanted to highlight this finding (section 3.1.1) before going into detail and describe sources and sinks of DFe and PFe. Here the explanation. We assume that particles in unfiltered seawater are identical to the particles collected by SAPS and just an incomplete dissolution of particles in unfiltered seawater should affect a similar result. All seawater samples were acidified with HNO_3 to pH 1.7 and stored for 1 year (supp. Text S1 line 36). Our data shows that this long and strong acidification re-dissolved 60% of particulate Fe from associated SAPS samples. The remaining 40% must belong to insoluble silicates. This is stated in the manuscript. The leachable particulate fraction from SAPS particles (LPFe) was determined by a 25% HAc leach for 2 hours at room temperature. It is wrong to believe that the HNO_3 and HAc leach are equal and deliver similar results. The pH of a 25% HAc solution is above 2.1 and applied for a short period of time. The HAc leach is usually strong enough to re-dissolve just surface scavenged, some biogenic trace metals and trace metals in carbonates (see any sequential extraction schemes for sediments). As shown in Table 2, on average less than 1% of the Fe belongs to the LP fraction (SAPS), the remaining 99% belong to the refractory fraction (which includes oxides, silicates, ect.). However, with respect to that the HNO_3 leach of unfiltered seawater must clearly also re-dissolve particulate trace metal oxides and amorphous substances. Thus comparing LPUn and LP makes no sense. In addition we have not introduced the refractory and the leachable particulate fraction of SAPS particles. We just compare the results from two particulate fractions that showed comparable results! However, we slightly modified

the sentence at line 161ff and replaced "refractory" by "insoluble" in line 171. We agree refractory is misleading since it does not rely on RPFe in SAPS samples.

-For the second point, presumably, they want to use the bottle-derived leachable particulate data because of higher depth resolution than SAPS samples, so this should be a separate argument than the one about the presence of refractory Fe and Al. Answer: Exactly, see explanation above.

-Table 2: are the LPFe, LPAl, LPMn columns derived from the SAPS LP or the bottle LPun? This should be specified in the caption. The units for the LP are specified as being in percent. But these values are all very smallâËŸA ËĞ Tmostly less than 5%, even for Mn, which contradict what was stated in the text in lines 161-163: "The LPUn corresponded to ca. 63_4% of the PFe, 83_11% of the PAl and 100_10% of the PMn fractions." Presumably, this discrepancy arises because the Table 2 values are SAPSbased (LP/P), whereas the values in the text are mixing and matching bottle-based LPun and SAPS-based totals. Is that correct? If this is the case, then this means that the bottle-based LPun are much higher than the SAPS-based LPs. One might expect this to some degree, since the LPun is acidified to a slightly lower pH (_1.7) and for a lot longer than the SAPS-based LPs (25% acetic acid should have a pH _ 2.1), but I wouldn't have expected the difference to be so big. This suggests that the particle population collected by the SAPS may be a subset of the particles accessed by the LPun method, and calls into question whether normalizing or comparing the bottle based LPun by/to the SAPS-based P is appropriate or meaningful. As further detailed below, I suggest that the authors do not mix bottle-based and SAPS-based parameters (i.e. they should not report or interpret LPun/P). Answer: As indicated at the end of the caption of Table 2, data corresponds to particulate data from SAPS. We added "SAPS" to the caption in line 827. With respect to the comparison of P and LPUn (comment of the reviewer). We argue in section 3.1.1 that LPUn is a subset of P, not the other way round. As stated earlier we try in section 3.1.1 not to compare both data sets, because this is to some extend meaningless (we do not know what did not dissolve in unfiltered

seawater), but it is without saying interesting that the both parameters LPUn and P show the same distribution in the water column and highlight the strong concentration gradients observed.

Section 3.1.2: this section needs to be considerably revised to account for the following: -Re. lithogenic source of suspended particles: The extremely good correlations of the PFe to PMn and PFe to PAl concentrations (Fig 3a,3b) do support their contention that the particles have a single lithogenic origin (line 197), since biogeochemical processing of the particles would be likely to affect Mn, Fe, and Al differently, and therefore cause more scatter in the data. However, the slopes of their relationships don't support this. The authors base the conclusion that the suspended particles have a lithogenic origin on the slope of the PFe to PMn data (68 mol Fe/mol Mn), which they say agrees well with a typical crustal ratio (they use 60 mol Fe/mol Mn from Wedepohl, which is close to 57 and 50 mol Fe/mol Mn for UCC and BCC crustal averages, respectively, reported by (Taylor and McLennan, 1995)). However, PMn is not usually a good crustal indicator, since Mn-oxides are frequently a large component of marine particulate Mn. For this reason, PAl or PTi are more frequently used to assess how lithogenic the PFe is. However, their PFe to PAl relationship (slope=1.251 mol Fe to mol Al) far exceeds the slope of typical crust (UCC Fe:Al=0.21; BCC Fe:Al=0.41 (Taylor and McLennan, 1995)). Since the sediment elemental ratios from the southern shelf (Table 3, line 199) are close to these crustal averages, this suggests a fairly large component of the PFe that is in excess of a lithogenic source. One can derive a PMn to PAl ratio from their data=PFe:PAl / PFe:PMn = 0.018 mol Mn/mol Al, which is also greater than typical crustal ratios (UCC Mn:Al=0.0037; BCC Mn:Al=0.0082). So just comparing their slopes to crustal ratios to Al, one would expect there to be a fair amount of nonlithogenic Fe and Mn assuming crustal averages are a reasonable approximation of the local sources (a plausible assumption given that the sediment elemental ratios are close to crustal averages). The conclusion that the suspended particles are primarily lithogenic is therefore not supported by their data. Answer: We do not agree with the reviewer. We argue that the strong correlation between P elements supports the existence of a

single source for suspended particles with lithogenic origin (line 197ff). And then we explain in the two following paragraphs why we do think so! As stated in the text the elemental ratio of P is higher as the elemental ratio of S (sediments) and the earth crust (now stated in line 198). We explain in detail that more biogenic Fe in the suspended particulate pool would lower the Fe/Mn ratio (line 205ff), lower than the sediment ratio of 51.1. This is clearly not the case! From line 208 on, we suggest that DFe was scavenged onto inorganic suspended particle surfaces. This process increases the Fe/Mn and Fe/Al ratio of suspended particles, and that is exactly what we observed. We are irritated that the reviewer did not mention the term scavenging a single time, even though this is the driving force for the elevated elemental ratios. In addition, we know that Mn is not a perfect sediment particle tracer and that titanium would be much better for that. However, the Fe/Mn ratio is a nice tracer to differentiate between biogenic and sediment/lithogenic particles!

-The slopes of Fe:Mn and Fe:Al of the LPun fractions are each about half of those for the total particulate fractions (Fig 3a-d). Comparing the leachable and total slopes, this suggests that particulate Fe is about half as labile as particulate Mn or particulate Al. This is much more consistent with their wholly SAPS-based %leachable data from Table 2 (average %leachable Fe < average %leachable Mn_average %leachable Al), than with their LPun/P from lines 161-163 (Fe<Al<Mn), another reason not to mix the bottle-based (LPun) and SAPS-based (LP, P) parameters. Answer: Figure 3c and 3d show the relationship of the leachable particulate elements of unfiltered seawater. The relationship is not as strongly pronounced as for P but exists, and this is an important finding of this study. We stated earlier in section 3.1.1 that on average 100 % of particulate Mn, 63% of particulate Fe and 83% of the particulate Al can be re-dissolved by the acidification of unfiltered seawater with HNO3. This interplay creates the reduced slopes in Fig. 3c and 3d. However, the relationship of LPUn and P is discussed in section 3.1.1 and not in section 3.1.2, the section the reviewer is discussing right now.

-To rule out data quality issues, the authors should report results from an external stan-

dard such as a standard reference material that was run alongside the digestion of the suspended particle leaches/digests. External standards in the supplements were only reported for seawater and for sediments, which used a different digestion procedure than the particles. Was there an external standard measured for the suspended particles to ensure that the digestion method was working accurately? Answer: Because of so many different data sets we forgot to include the crm values for SAPS particle digests. We analysed two crm's, TORT-2 and NIST 1573a. Values are now stated in the supplementary Text S1. A link to this data is provided in the main text body, section 2.2 line 134ff.

Section 3.1.3 -Re. meltwater source of LPunFe: is there a relationship between LPunFe and salinity? A scatter plot of these parameters would be evidence of a meltwater source of LPunFe. Answer: We included a graph in the supplementary material (now Fig. S3) illustrating the relationship between salinity vs. dissolved (DFe) and leachable particulate Fe (LPUnFe).

-Lines 253-256: PFe/PAl in krill fecal pellets (0.48) is much lower than PFe/PAl in suspended particles (1.25âËŸAЁ TFig 3b), though it is relatively close to the sediment ratios from the southern shelf. This suggests that the suspended particles have an additional source of PFe compared to krill or sediments. Answer: On the other hand, the PFe/PMn ratio is almost the same. Schmidt et al. [2016] showed that at some locations on the shelf the content of lithogenic particles exceeds more than 50% of the krill stomach content (Figure 1H). However, we added a possible explanation for this discrepancy (line 256ff).

Section 3.2.1 -The authors should acknowedge that the sediment core data were taken on the nonproductive, southern side of the island, whereas the water column data were taken from the productive, northern side of the island, so benthic fluxes calculated from porewater profiles could be rather disconnected to the measured DFe. Answer: A sentence was included that refers now to benthic flux differences between the northern and southern side of the island (line 291ff). However, benthic fluxes into bottom waters

are irrelevant for our water column fluxes calculations (see supplementary data).

-The authors calculate a vertical supply of DFe to surface mixed layers by assuming a vertical diffusivity and vertical advection (upwelling) used by De Jong et al. 2012 for the region downstream of the Antarctic Peninsula. But later in section 3.4.2, they discuss a study by Tagliabue et al. 2014 modelling DFe supply for South Georgia, in which Ekman *downwelling* (not upwelling!) prevailed, removing DFe from the surface, rather than supplying it. I can appreciate that there's uncertainty in the estimate of vertical advection, but they should probably pick a sign and stick to it! Answer: Both regions, the shelf and bloom region, are separated by 1.250km and have to face different oceanographic settings. This is stated in the manuscript section 3.4, line 448. Tagliabue et al. 2014 modeled the dissolved Fe supply over the entire Southern Ocean. This model, which is based on observational data, suggests that the main Fe source south of the Polar Front (PF) is Eckman upwelling of nutritious deep waters. North of the Polar Front (South Georgia is located north of the PF) the advective term of vertical missing is negative (downwelling) and the deep winter entrainment is responsible for a large fraction of primary productivity found. However, near an island with shallow topography this scenario is entirely different, and that is true even north of the PF. As already pointed out, internal waves that cross a shallow shelf produce large turbulences, which supports positive vertical advective mixing of nutritious deep waters into the surface layer [Kurapov et al., 2010; Moore, 2000; Wolanski and Delesalle, 1995]. This turbulences are even strong enough to bring sediment particles in re-suspension (what we see on the shelf). Because of that we applied in addition to the diffusive mixing term also the advective term. The Kz and w terms were inferred from de Jong et al. (2012) collected near the Antarctic Peninsula. For our opinion these numbers reflect the advective and diffusive force best for a shallow shelf in the Southern Ocean. However, for clarification we replaced the term upwelling by "advective" velocity and Eckman "transport".

Section 3.3: there are some problems with the proposed flowpaths: -Line 384: I do not understand the proposed advective pathway: the authors reference "Fig. 1; #11/12

via #13 to #14" as a NE-SW transect. First, these stations do not define a NE-SW transect. Second, what is the order of the proposed advective pathway? Is it 13 to 14 to 11/12, even though 13 is actually further from the island than 14? Is 13 shallower than 14 (Fig 4) despite being further offshore (Fig 1)? Answer: We thank the reviewer for their comment. Indeed the pathway is from St. 14 near onshore via St.13 to St. 11/12 offshore. We changed that in the text and mention that the pathway is in E-W direction. Because stations were sampled one after another and are located on an almost linear line from 25 km away from the coast to 70 km offshore we decided to choose this transect. On the other hand this transect represents the only almost linear transect from onshore to offshore with more than two stations, as mentioned later by the reviewer. However, shelf topography of South Georgia is highly variable and varies usually between 200 and 250 m depth (Fig. 2). Nevertheless, locations with a shallower water columns are possible, as we found this for site #13 with ca. 133 m depth. However, this is what we found!

-Presuming that the authors are assuming that #11/12 is the offshore end of the flow-path, this is inconsistent with DMn, which is higher at 11/12 compared to 13 and 14. It's no wonder that they were not able to fit an exponential function to the DFe data (lines 409-410). If this isn't an advective flowpath, the good exponential fit to the LPUnFe data may be a coincidence, or at least unrelated to offshore transport. Further, the "exponential decrease" in PFe was based on a 2 point fit! No wonder R2=1! This should be removed. Answer: We were also surprised that LPUnFe concentrations decreased exponentially with increasing distance to the coast so well. However, suspended sediment particles sink quickly when turbulent mixing decreases. Due to this we expect that shortly after the shelf break was passed most sediment particles are vertically exported. This is in agreement with the literature. So, chances are very slim that these sediment particles below the mixing layer between 200 m and 400 m contribute significantly to the Fe budget approx. 1,250 km downstream the island. Particles mainly sink! We mention this here because the reviewer said earlier we introduced an PFe fraction here that was not taken into account for the later budget. However, to clarify this we

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added a sentence (line 420ff). However, this is different for DFe. This Fe fraction can vertically rise, driven by vertical diffusive mixing and strong concentration gradients. Anyway, for a more robust horizontal flux calculation we decided to take the entire DFe data into account. Because of the large variability of the data set and an overall short transect length (max. 80 km away from the coast; de Jong et al. (2012) showed data 3500 km downstream the source region) we could not fit a line through the data, which is needed to calculate the scale length and Kh (horizontal diffusivity (de Jong et al. (2012)). Sorry, this is what we found and I think this is the best way to handle the data carefully.

Section 3.4: budget -As noted above, there is a discrepancy in the manuscript about whether this region is downwelling, as assumed here for the budget, or upwelling, as assumed in section 3.2.1âËŸAËĞ Tsediment DFe flux. Answer: This has been discussed in detail earlier. However, the reviewer must have not understand that the vertical advective mixing is different at the Fe source region and the region of elevated photosynthetic activity [Tagliabue et al., 2014]). However, we included this now in the text section 3.2.1, line 328ff) and leave the discussion open if the advective term should be used or not.

-In the manuscript, an overall vertical loss of DFe of -0.0025 umol DFe/m2/d is assumed (line 465), but in Figure 8, a vertical supply of +0.009 umol DFe/m2/d is indicated. Answer: As pointed out in Figure 8 the vertical input from below is the sum of diffusive (diapycnal = 0.0003 umol m-2 d-1), advective (Eckman = -0.0028 umol m-2 d-1) and deep winter mixing (entrainment = 0.011 umol m-2 d-1). With respect to the fourth and fifth decimal place we calculate a vertical flux from below ca. 0.009 umol m-2 d-1.

-Given the poor (lack of) constraints on both horizontal and vertical supply of DFe, the suggestion that there is a mismatch in the supply and demand that is filled by advecting biogenic Fe is a nice hypothesis, but rather speculative, both the size of the term, and the nature of it. The size of the flux is not constrained, since they did not have local estimates of horizontal or vertical supply. Re. the nature of their proposed flux: their

PFe:PAl ratio from earlier in the manuscript does suggest excess particulate Fe over lithogenic, so this is at least consistent with an available biogenic Fe pool on the shelf, but is also consistent with Fe oxyhydroxides (i.e., authigenic, not biogenic particulate Fe). Answer: Our observational data from the shelf indicates that the South Georgia shelf represent a strong DFe source that supports elevated primary production and the growth of larger organism (located at a higher trophic level) on the shelf. Fe fluxes calculated for the bloom region rely on validated literature values (as this is described in the text extensively). Tagliabue et al. (2016) and Chance et al. [2015] showed that aeolian dust deposition, horizontal and vertical Fe input delivers enough DFe to support phytoplankton growth in most low productivity waters in the Southern Ocean. However, with respect to the estimated Fe requirements of the phytoplankton community in high productivity waters, such as downstream the island of South Georgia, these sources are two low to explain the observed growth. So, DFe needs to come from somewhere! Tagliabue et al. (2016) argued that mainly the entrainment of DFe during the winter drives the phytoplankton growth. However, winter entrainment is a one-time event in the winter and increases the DFe pool in surface waters temporarely. He suggests that most of this DFe is incorporated in phytoplankton cells (biogenic Fe by luxury Fe uptake) in the early time of the growing season. This biogenic is then internally recycled in the surface mixed layer during the entire growing season. We argue that this on-time shot of DFe is not enough in high productivity waters. We suggest that in addition Fe enriched biogenic particles/phytoplankton cells are formed by luxury Fe uptake on the shelf and that these Fe enriched biogenic particles are internally recycled when water masses are transported downstream away from the island. See section 3.4.5 line 516ff.

Miscellaneous: -Lines 104-107: I assume that unfiltered samples were also collected from the tow fish in JR247, since LPun values are plotted for this cruise in figure 6. Please add this to this section. Answer: We added "Unfiltered surface seawater samples were collected and dispensed into acid washed 125 mL LDPE bottles." line 107ff.

-Line 231-2: Fig 6a shows that surface concentrations of 0.3 _M LPunFe are quite

common on the shelf, but 22 uM doesn't show up in this figure, so referring to it here is not that helpful. Answer: We removed the figure citation. However, LPUnFe concentrations are usually lower around 2.2 uM near the shore and <10 nM after the shelf break. We decided to not us a log scaling just for one data point.

Typos: Line 106: "cartridge filter", not "filer" Line 178: "where", not "were" Line 251: "ascent" not "ascend" Line 415: add "there was" to "...that *there was* a limited transfer..." Line 459: "Tagliabue" not "Tagliabu" Answer: Changed as suggested.

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