

Dear Editor,

We thank all three reviewers for the constructive comments which have helped to improve the manuscript.

We did not provide responses to the opening questions as we had the feeling that the opening questions were repeated within the specific comments. As such we did answer the specific comments.

Overall, we have made clear the differences between surface and deep sections and added new figures. We added a figure with the correlation between dAl and Si. We have added linking sentences between sections. The presence of BSi and pAl is of major importance and we are confident that both manuscripts are being submitted before the deadline.

Below you will find all the answers to the specific comments and technical corrections of all three referees.

Kind regards,

Jan-Lukas Menzel Barraqueta on behalf of all co-authors

Anonymous Referee 1:

Specific comments:

- *As mentioned above, authors discussed about the source and sink of the dissolved Al with mainly controlled by remineralization of the biogenic particles because of the positive correlation of the Si and Al concentrations in the water column. However, there is no plots between these parameters in this manuscript. Also, it is not clear that the correlation of these parameters was obtained within the same water depths (Euphotic zone in Section 3.2.2 or water column in Section 3.3.1) and/or same water masses (since it is interesting that the advected water mass (i.e. MOW) showed the different correlation of Si/Al.) Need to clarify this information in this manuscript.*

→ We have clarified the issues as they were also raised by referee 2 and 3, and included this information in the manuscript. We have included a new figure (S5) showing the correlation between dAl and Si for the whole section. Now section 3.2.2 and 3.3.1 reads as follow:

age 7, line 9-35, page 8 1-9: Section 3.2.2: Biogenic opal production and biogenic particles play an important role in the removal of dAl in the surface ocean as a result of the high particle surface affinity of dAl (Moran and Moore, 1988b). Removal of dAl by particles therefore represents a mechanism which reduces dAl and increases pAl concentrations in surface waters (Moran and Moore, 1988a). During the GEOVIDE cruise, Chl a concentrations increased from the NAST (0.2 to 0.6 mg m⁻³), via the NADR (0.5 to 1.3 mg m⁻³) to the SANA region (up to 5.5 mg m⁻³ at station 61) (Tonnard et al., b, special issue). Along the GEOVIDE transect, diatoms were the dominant phytoplankton taxa (>40% of total phytoplankton community) in the ENAB, IrB, and LB while coccolithophorids were dominant (50% of total phytoplankton community) in the IcB (Tonnard et al., b, special issue). Elevated bSi and pAl concentrations and low dAl concentrations coincided with diatom dominated phytoplankton communities for the ENAB, IrB, and LB (Fig. 2). In contrast, elevated dAl and low bSi and pAl concentrations coincided with coccolithophore dominated phytoplankton communities for the IcB (Fig. 2). The latter could suggest a preferential scavenging of dAl by diatoms rather than by coccolithophorids. This conclusion is supported by increased pAl to dAl ratios where surface waters were dominated by diatoms (Fig. 5), as possibly a consequence of active dAl incorporation into siliceous shells (Gehlen et al., 2002) or scavenging of dAl onto biogenic opal (Moran and Moore, 1988a). One way ANOVA analysis was performed for the pAl to dAl ratio in the surface waters (>50 m) in each of the four basins which showed strong dAl to Si(OH)₄ correlations with depth (LB, IrB, IcB and ENAB, see section 3.3.1). The ANOVA test showed significant differences (at the p <0.01 level) between basins [F (3, 38) = 7.9, p=0.0003]. Post hoc comparisons using the Tukey HSD test showed significant differences between the LB and IcB (p <0.01) and ENAB (p <0.05), and between the IrB and IcB (p<0.01). No significant difference was observed between the IrB and LB and ENAB (p =0.86 and 0.28, respectively), and between ENAB and IcB (p=0.3). Taken together, our results indicate that when diatoms were abundant, as in the IrB and LB (>60% of total phytoplankton community, Tonnard et al., b, special issue), the ratio between pAl and dAl significantly

increases due to dAl sorption onto biogenic opal surfaces and the transfer of Al from the dissolved to the particulate fraction. Similar observations have been reported for laboratory studies and in the field where dAl decreased as a function of diatom growth and/or the presence of enhanced quantities of biogenic particles (Hydes, 1979; Kremling, 1985; Kremling and Hydes, 1988; Measures et al., 1986; Measures et al., 1984; Orians and Bruland, 1986; Stoffyn, 1979; van Bennekom, 1981). In addition, the transfer from dAl to pAl has been observed in coastal regions (Brown et al., 2010; Moran and Moore, 1988a) and the North Atlantic (Barrett et al., 2015). Considering the low aerosol deposition, reduced fluvial inputs into the tested basins (ENAB, IcB, IrB and LB), and elevated levels of bSi, we conclude that the observed differences in pAl to dAl ratios were mainly related to the of diatom abundance.

Page 11, line 6-35, page 12, line 1-4: Section 3.3.1: In the remote oligotrophic regions of the North Atlantic Ocean with enhanced Saharan dust inputs, dAl shows enhanced surface water concentrations with depletion at depth (Measures et al., 2015), typical for a scavenged type element (Bruland et al., 2014). A scavenged type distribution for dAl has also been described for the Pacific Ocean (Orians and Bruland, 1985). In contrast, a nutrient type depth distribution of dAl has been reported for the Arctic Ocean (Middag et al., 2009), Mediterranean Sea (Hydes et al., 1988; Rolison et al., 2015b), North Atlantic (40°-50°N) (Barrett et al., 2012; Measures et al., 2008) and high latitude North Atlantic (Middag et al., 2015b) and these distributions coincided with strong correlations between dAl and Si(OH)₄ (Hydes et al., 1988; Middag et al., 2015b; Middag et al., 2009; Rolison et al., 2015b). Dissolved Al is thought to be removed from surface waters onto particle surfaces (Moore and Millward, 1984; Orians and Bruland, 1985), including diatom cells (Gehlen et al., 2002) and subsequently released at depth during the recycling of biogenic particles and desorption from non-biogenic particles. In our study region, diatoms dominate the phytoplankton communities at the early stage of the spring bloom (Brown et al., 2003), and are an important producer of biogenic silica (bSi) (Nelson et al., 1995). This, along with other biogenic particles, is a main carrier for scavenged Al (Moran and Moore, 1988b; Stoffyn, 1979). Elevated bSi concentrations and associated high export rates of bSi were measured using in situ pumps in the ENAB (up to 1.19 μM bSi), and in the IrB and LB (up to 4.27 and 4.63 μM bSi, respectively) (Lemaitre et al., special issue). Dissolved Al and Si(OH)₄ displayed strong correlations (full dataset R²=0.56, Fig. S5) with depth in all basins (ENAB, IcB, IrB and LB) (R²>0.76), except in the IB (R²= 0.2) which featured Al enrichment from the MOW (See section 3.4.1), the Tagus estuary and the Iberian shelf/margin (see section 3.2.1 and 3.4.2). The large production of opal and other biogenic particles (e.g. CaCO₃ from coccolithophorids) (Lemaitre et al., this issue), the strong correlation between dAl and Si(OH)₄ with depth, and the increase in dAl concentrations with depth (Fig. 7a and b, see section 3.3) suggest that in the water column the net remineralization of dAl from particles was larger than the net removal of dAl from scavenging. However, it should be noted that dAl removal by diatom production is not necessarily the only reason for the nutrient type distribution. Whilst we did not observed enhanced pAl concentrations where coccolithophorids were dominant (e.g. IcB; section 3.2.2), we assume that also other biogenic and non-biogenic particles (e.g. CaCO₃, organic carbon, lithogenic particles, zooplankton fecal pellets) will contribute to the vertical export

of surface dAl into the deep ocean. Advection and mixing of water masses with different pre-formed dAl concentrations will also influence water column distributions of dAl, as observed for water masses with low dAl concentrations (e.g. DSOW) in comparison with overlying waters (e.g. ISOW; see section 3.4.2). Enhanced sediment resuspension can furthermore add dAl to bottom waters (See section 3.4.2).

- *Abstract: Line 19. “: : . and removal by phytoplankton.” Dissolved Al is not a bioactive metal (in the current knowledge), thus this sentence is probably misleading readers. This sentence should be rephrased with “removal by biogenic particles (i.e. phytoplankton)”.*

→ We have rephrased the sentence as suggested.

Page 1, line 18-19: Surface water dAl concentrations were low (median of 2.5 nM) due to low aerosol deposition and removal by biogenic particles (i.e. phytoplankton cells).

- *Section 2.2 Dissolved Al analysis. (Page 4, first paragraph) Authors described the analytical method with the loading time (second) only. It is very helpful for readers to add the volume or loading time and the flowrate in this sentence. For example. “The loading time was adjusted to 120 s (flowrate ? mL/sec, or ?? mL) and was extended up to 180 s (flowrate ? mL/sec, or ? mL) for samples: : :”. Same as the rinsing volume and elution volume.*

→ We have included the flowrates.

Page 4, line 6-10: The loading time was adjusted to 120 s (2.5 mL/min) and was extended up to 180 s for samples with low dAl concentrations (<2 nM). After sample loading, the column was rinsed for 70 s (2.5 mL/min) with deionised water (18.2 MΩ cm⁻¹, Milli-Q, Millipore) to remove the seawater matrix major seawater anions that interfere with analysis. Subsequently, the preconcentrated dAl was eluted (120 s, 0.6 mL/min).....

- *Page 5 Line 8. “The blank was subtracted from: : :”. Authors should write the blank value here. Is the value 0.013nM??*

→ Done. No, the average blank was 0.23 nM ± 0.1 nM (n=28)

Page 4, line 25-26: The total blank (Mean = 0.23 nM ± 0.1 nM; n = 28) was subtracted from the results obtained.

- *Page 7. Line 7. Did authors calculate the averaged dAl value (3.1nM) including Station 1, 2, 4? The dAl value at Stations 1, 2, 4 were quite high (>20nM) in the Figure 2. Authors should specify the station number in this sentence. Add the station information for ENAB and IcB as well.*

→You are right. In fact the average number given does not include stations 1, 2, and 4. We have clarified the information.

Page 6, line 15-18: Average surface dAl concentrations decreased from 3.3 ± 1.7 nM (n=5) in the IB (Stations 1, 2, and 4 are excluded due to elevated dAl concentrations due additional inputs from the Tagus estuary) to 3.2 ± 0.8 nM (n=4) in the ENAB (Stations 21 to 26) and 2.8 ± 1.2 nM (n=5) in the IcB (Stations 29 to 38),

- Page 8. Line 14. *The correlation here was referred to Section 3.3.1. However, this section is describing for surface/Euphotic zone, not deeper water. Need to specify or rephrase it.*

→We have clarified this.

Page 7, line 26-28: One way ANOVA analysis was performed for the pAl to dAl ratio in the surface waters (>50 m) in each of the four basins which showed strong dAl to Si(OH)₄ correlations with depth (LB, IrB, IcB and ENAB, see section 3.3.1).

Technical corrections:

- Page 5 Line 19. “The Fisher-based test”, is this mean “Fisher’s exact test”?

→You are right. With “Fisher-based test” we meant “Fisher’s exact test”. We have changed the name.

Page 5 line 4: The Fisher’s exact test was used for.....

- Page 6. Line 13. *“Subarctic North Atlantic Artic”. Is this “Arctic”?*

→The Irminger Basin and Labrador Basin (d and e on figure 1) are part of the biogeochemical province SANARCT. It is not the Arctic but the latter biogeochemical province includes regions which are in the Arctic and which share similar physical and ecological characteristics. However, we have remained this biogeochemical province to SANA (Subarctic North Atlantic) throughout the manuscript.

- Page 6. Line 25 and 26. *The numbers should be written as (iv) and (v), not (iii) and (iiii).*

→Changed.

- *Page 7. Line 6. "... and 30.99nM (Station 2)." Figure 2 shows the highest dAl value was observed at Station 4. Is this station number 4 in this sentence?*

→Indeed, there is a mistake on figure 2. The highest dAl concentration corresponds to station 2. Corrected. Station 2 is now shown in figure 3.

- *Page 9. Line 24. High riverine dAl signal was observed previously in the Bay of Bengal (see the reference of Grand et al (2015)).*

→We have included the suggested reference.

Page 9, line 3-6: Our results indicate that a fraction of riverine dAl can be advected offshore, as observed previously in the Bay of Bengal (Grand et al., 2015),....

- *Figure 1. Station number in Figure 1b would help reader to understand the figure better. Figure caption "C: Atlantic Artic (ARCT)" should show "C: Subarctic North Atlantic Arctic (SANRCT)" in order to corresponding the text.*

→We have changed figure 1 based on the review by Dr Resing. We have added station numbers on figure 1b.

- *Figure 2. Figure caption. ": : .Green: NAST (A in Fig 1); Orange: NADR (B in Fig 1); SANRCT (C in Fig 1).."*

→We have redone figure 2. Therefore the caption has been changed accordingly.

- *Figure 5. It would be better to show "Greenland" in the Figure.*

→We have added "Greenland" on the three plots.

- *Figure 6. (A) why the station 2 was highlighted with red color? (C) It would be better to put the station numbers, black dots and the geographic labels "Newfoundland" etc..like Fig 6a. In order to help reader, modify the figure 6 with focusing shallower depth or add separate figures (see above).*

→It was a mistake. Suggested changes done.

Referee 2 (Rob Middag)

Specific comments:

- *Page 3 line 28 why were two different filters and pore sizes used and is there a difference between them and the Al fraction analysed?*

→ During some cruises we filtered the samples with capsule filters (Sartobran 300, Sartorius) and sometimes the samples were filtered with the filters used to collect the particulate trace metals (Supor®, Pall Gelman). No, there is no difference and this approach is common on a lot of the GEOTRACES cruises as water use is optimized to allow collection of filtered seawater and particulate metals. Both filtered fractions are considered as the dissolved fraction (www.geotraces.org/).

- *Page 4 line 22 Brown and Bruland used a 4 M buffer. Is there a reason for changing the concentration?*

→ Indeed, this is a slight modification of the method published by Brown and Bruland. We have added a sentence to clarify this. The important aspect was to reach an optimal reaction pH between 5.2 and 5.6 which was also reached with the 2M buffer. Another advantage is that you can prepare a single buffer for the Al analysis instead of having to prepare two different buffers.

Page 4, line 2-4: A slight modification of the method published by Brown and Bruland (2008) is the use of a 2 M ammonium acetate buffer (UpA, Romil), instead of 4 M, in the reaction stream.

- *Page 5 line 5 What is the effect on the sample pH when adding 3 times the required amount of buffer and the subsequent pre-concentration? I find the blank determination slightly odd. If I understand it correctly, 'regular' samples are buffered online, but for the blank determination a sample was buffered offline with varying amounts of buffer. Additionally an acidified MQ sample was analysed without buffer (so I assume the system had to be modified, i.e. the buffer line was removed?). Wouldn't it be far easier and more representative to analyse an acidified MQ sample as a regular sample?
With the current approach, any blank resulting from the online buffering (if any) is not accounted for.*

→ Adding three times the required buffer will elevate the sample pH upon preconcentration. However, in the improved method published (Brown and Bruland, 2008) it is shown (Fig. 2) that at a pH above 5 the effect of sample pH on column retention of Al is negligible. Indeed, the buffer line is “closed” for the blank determinations. The manifold blank was determined running an acidified seawater samples (not an acidified MQ sample) without buffering. Running an acidified MQ sample would be different in terms of sample matrix. Yes, it could be far easier to

proceed as suggested, but overall it looks that the differences which arise from our procedure are relatively small or absent as the analysis were certified by the run of GEOTRACES reference seawater and internal standards. However, for future analysis we will bear in mind this recommendation.

- *Line 8 the average blank and standard deviation should be reported*

→Done.

The total blank (Mean = 0.23 nM ± 0.1 nM; n = 28) was subtracted from the results obtained

- *Line 17 I would say similar analytical techniques in different labs and cite the paper (Rolison et al., 2015) for the other dataset. Here it should probably also be pointed out that one dataset was analysed shipboard, the other after storage for some time in a shore based lab and that there appears to be no difference between the approaches (At the GEOTRACES website there is a cautionary note accompanying the SAFe reference sample results warning there could be Al contamination from bottle caps during storage).*

→We have changed this information and clarified the procedure.

Page 4, line 34-35 and page 5 lines 1-3: the GEOTRACES section GA04N (Station 1, 39.73

°N, 14.17 °W) (Rolison et al., 2015). Dissolved Al was analysed, based on the same method, using similar analytical techniques in different laboratories. It is noteworthy that dAl samples for GA04N were analysed on-board ship and for GA01 after prolonged storage at a later date. Samples analysed for GA01 were stored up right in order to minimize any contact and potential contamination arising from the polypropylene caps.

- *Page 7 line 25 room for clarification here; in the previous paragraph decreasing concentrations from eastern to western basins were described, so I guess the point here is that there is no significant difference? Also the decrease would not only be expected in the east-west direction, but also in the south north direction (away from Saharan dust source).*

→Clarifications made. We have added a sentence clarifying that within errors no significant difference was observed. We have also added that atmospheric aerosol loading decreases in a westward and northward direction from the main aerosol source (e.g. Sahara).

Page 6, line 21-22: However, within analytical errors, no significant difference was observed between the basins.

Page 6, line 29-30: In the North Atlantic atmospheric aerosol loading declined in a westward and northward direction, i.e. with increasing distance from the major African dust source regions....

- *Section 3.2.2 What stations are used in the calculations? Probably the coastal stations should not be used here as particle concentrations and compositions here are*

influenced by continental and sediment sources as discussed in subsequent sections. Additionally, this section warrants some further discussion on the use of Al as a dust tracer, as this study implies the dissolved Al concentration is not only dependent on atmospheric deposition, but also the presence of diatoms.

→The following point was addressed also by referee 1. We have added the stations numbers. See referee report 1. Yes, you are right. We are preparing a manuscript on the role of dAl as a dust tracer using surface mixed layer dAl concentrations from GEOVIDE, GA06, GA08, and GA10 cruise. Therefore we have not included a discussion on this topic.

- *Section 3.2.3 I'm not 100% convinced based on the current discussion the elevated Al is associated with river outflow; how can one be sure e.g. it is not all wet deposition or that sediment resuspension on the shelf doesn't play a role too (after all, a sediment resuspension source is argued as a significant source in section 3.4.2). Was there a correlation between dAl and salinity or other tracers of fluvial input (as for example observed in the Drake Passage for Al input associated with land run off (Middag et al., 2012))? The authors have the data to discuss this in more depth and to discuss the importance of fluvial input vs deposition and resuspension (the Al could be effectively removed but later re-suspended as suggested for the Californian shelf for Fe by Bruland and co workers).*

→We extended the discussion about this topic. Based on the data we have, the resuspension of shelf sediments is unlikely to be an important source to surface waters. We have added a new figure in the SI which shows that sediment resuspension is a source for bottom waters, but not a source for surface waters. Wet deposition is also unlikely to be the main source of dAl as the shape of the salinity profiles for stations 1 to 4 and for station 11 are different. Thus, we still consider that the river input is the main source for the elevated dAl concentrations observed.

Page 8, line 21-35, page 9 line 1. Possible sources which could explain the elevated surface dAl concentrations are shelf sediment resuspension, wet deposition, and riverine inputs. Shelf sediment resuspension is unlikely to be the reason for the elevated dAl concentrations as deep profiles for stations 2 and 4 (Fig. S1) showed that the elevated levels of dAl observed in bottom waters were not a source for surface waters since minimum dAl values were observed between maximum surface and deep dAl values. Salinity profiles for GEOVIDE showed salinity minima (<35) in surface water for stations 1, 2, and 4 (Fig. S2), indicating a freshwater source. No evidence of freshwater input was observed at station 11, located just west of station of 1. Plausible explanations for the observed distribution in salinity are therefore wet deposition and/or river inputs. Wet deposition events were registered between stations 1 and 4 (Shelley et al., 2017a). Yet, the shape of the salinity profiles of stations 1 to 4 seem unlikely to have been caused solely by recent wet deposition as the differences in salinity were observed up to a depth of 45 m. GA01 ship's ADCP data (Fig. S3) showed that surface waters near the Iberian Peninsula flowed in a northward direction. Therefore, we suggest that the additional source of dAl to surface waters originated from the Tagus estuary located approximately 175 km south from stations 1, 2 and 4. Elevated concentrations of dissolved Fe in the Tagus outflow and strong correlations for salinity against dAl and dFe (Tonnard et al., 2018) observed during the GA01 cruise supports a riverine source of dAl.

- *Section 3.2.4 I find the argumentation in this section a bit shaky as detailed below. I do not disagree with the point reached, but the argumentation needs to be improved
Line 7-10 it is probably worthwhile to mention these endmember estimates are conservative for dAl as they do not incorporate any Al scavenging or precipitation. Is that also the reason for pooling the dAl and pAl later in the paragraph when discussing endmembers?*

→ We have clarified that the endmember estimates for dAl are conservative. No, there was no specific reason.

- *Page 10 Line 10 I do not see how samples from a fjord and an iceberg are representative for glacial runoff and sea ice melt. Sea ice melt will be completely different (see also next comment) and the concentrations in the fjord will not only depend on run-off, but also the interaction with sediments and (biogenic) particles in the water column*

→ Biogenic particles are not a major issue for low-salinity samples from inner-fjord environments, these environments are invariably light limited, low productivity environments (chlorophyll a in the region where these samples were collected was < 0.25 mg L⁻¹), but of course fine sediment is also a source of trace elements in these near-shore environments which is difficult to ‘uncouple’ from freshwater input. The interaction with sediments is a point we now address. We better distinguish between these sources in R1 and state the inevitable limitation that overlap between these sources makes it difficult to distinguish between them, but note that the high pAl signal we observed requires a terrestrially derived source.

- *Line 12-13 why would sea ice Al concentrations be more similar to ice berg than glacial run off? Ice bergs were once part of the glaciers, so wouldn't one expect that ice bergs and glaciers are quite similar and sea ice very different from those two?*

→ Meltwater/runoff acquires, by marine standards, massive concentrations of dAl and pAl in pro-glacial streams or in shallow coastal areas around Greenland where sediment plumes are present throughout the year. Thus the pAl signal ‘delivered’ into the marine environment is much higher than that arising in ice melt itself. Hence we expect that the runoff derived signal will have a much higher pAl and dAl content than either ice-melt or sea-ice melt due to the greater sediment-water interaction, this is verified by considering the range of dAl and dFe data reported in prior work for these sources.

- *Line 14-15 the comparison between total dissolvable and dAl+pAl is not valid in my opinion, the total dissolvable is a ‘gentle leach’ at pH 1.8 whereas the pAl analysis is a complete destruction.*

→ We now explicitly acknowledge in the text the difference between pAl and tdAl, but this does not change our finding that the relative contribution of different sources which is the basic point made.

Page 9 line 21-35 and page 10 line 1-8: Freshwater endmembers (salinity 0) for Al were determined from linear regressions between dAl, pAl and salinity for the eastern stations (49, 53, 56 and 60; dAl 60.5 ± 9.9 nM and pAl 773.7 ± 125.6 nM) and western stations (61, 63 and 64; dAl 6.2 ± 1.2 nM and pAl 675.1 ± 124.7 nM) on the Greenland shelf (Table 1). These endmember estimates will be considered conservative as they do not incorporate Al scavenging processes. To gain some insight into what sources may have

contributed most strongly to our high Al signals in surface waters off the Greenland shelf, we analysed a collection of iceberg and fjord samples from west Greenland. Mean total dissolvable Al (unfiltered) iceberg and fjord concentrations were 55 ± 2 nM and 12.8 ± 6 μ M, respectively (Table S1). Freshwater Al endmembers (dAl + pAl) derived from our shelf stations were an order of magnitude higher than the mean tdAl measured in iceberg samples. tdAl must by definition be < pAl due to the weaker leaching procedure applied. Yet given the large difference, the Al values off Greenland appear to be related to the input from terrestrial runoff enriched with glacially derived sediment, with this enrichment occurring either downstream of glaciers in pro-glacial environments or in near-shore environments where sediment plumes can result in high trace element concentrations throughout the year (Hopwood et al., 2016). This is consistent with a similar elevated Fe signal on the Greenland shelf. Similar observations of elevated Al were made downstream of a glacier catchment in Cumberland Bay (Schlosser et al., 2017), and attributed to suspended glacial flour as the main source for enhanced pAl concentrations. An alternative low salinity dAl signal could come from sea-ice, which contributes a total freshwater input to the EGC approximately equal to that of terrestrial runoff (Sutherland et al., 2009). Whilst sea-ice dAl concentrations are not available for this study/region, Lannuzel et al. (2011) reported median dAl and pAl concentrations in sea ice (pack ice) of 2.6 and 10.7 nmol L⁻¹. We therefore anticipate that local ice-melt (from sea-ice and icebergs) formed a minor contributor to the shelf dAl signal compared to terrestrial runoff.

- *section 3.3 title of this section could be improved, section 3.2 has an informative title and deals with surface water, what is this section about and what distinguishes it from section 3.2 (as that section also deals with spatial distributions)*

→ We have modified the title of section three. Section 3.3 deals with the vertical dAl distribution during the GEOVIDE cruise while section 3.2 deals with the surface dAl distribution.

Page 10 line 10: Overview of the water column distribution of dAl

- *Section 3.3.1 I think this section should be better linked to section 3.2.2. Is there anything that can be learned from (changes in) the particulate phase in deeper waters in this region? And an increase with depth is not only related to local (vertical) processes, but also processes during advection of deep water masses and mixing of water masses with different pre-formed concentrations. The latter should also be explored/discussed as a potential driving factor behind the observed correlations. Notably in the next section, sediment re-suspension is discussed which also lead to increased concentrations at depth so a better linking of section 3.3.1 with 3.4.2 is also warranted.*

→ We have added some sentences linking sections 3.2.2 and 3.4.2 with section 3.3.1. We have added the potential factor of advection/mixing and sediment resuspension in controlling the dAl water column distribution. We are not sure about the point made with “anything that can be learned from the particulate phase in deeper waters in this region”. We have added a plot in the comments to reviewer 3 showing the distribution of the pAl to dAl ratio with depth. However, it is to note that the focus of this paper is dAl and not

the particulate phase. The particulate phase will be discussed in Gourain et al., this issue.

Page 12 line 1-5: Advection and mixing of water masses with different pre-formed dAl concentrations will also influence water column distributions of dAl, as observed for water masses with low dAl concentrations (e.g. DSOW) in comparison with overlying waters (e.g. ISOW; see section 3.4.2). Enhanced sediment resuspension can furthermore add dAl to bottom waters (See section 3.4.2).

- *Section 3.4.2 Page 13 Line 4-5 how does a positive correlation between pAl and salinity indicate the salty MOW is depleted in pAl?*

→ I guess, you wanted to mentioned section 3.4.1. The point we wanted to make was that pAl concentrations seem depleted in comparison with dAl concentrations. We have rephrased the sentence to avoid confusion.

Page 12 line 20-22: The linear correlation between dAl and pAl and salinity, and the shape of the vertical distribution of dAl at station 11 (Fig. S4) indicate that the MOW is highly enriched in dAl and represents a major source of dAl to mid-depth waters in the North Atlantic.

- *Line 9 Do we need another tracer for MOW? Conventional tracers such as salinity that are far easier to measure than Al seem to work well, so what is the benefit of the relatively expensive and challenging parameter Al?*

→ No, of course not. Salinity is an “easy” to measure parameter which perfectly traces the MOW. The reason behind measuring Al is not the use as a tracer of water masses (Although this has been acknowledge before (Measures and Edmond, 1990)), it is the importance of Al as an indicator for lithogenic inputs. As such, it is included as key trace metal in the GEOTRACES project.

- *Line 28 were there more stations on the shelf without elevated concentrations? Currently not clear.*

→ Yes. Station 56 over the Greenland shelf did not present any significant elevated dAl concentrations in the most bottom sample.

- *Page 14 line 3-8 not directly clear station 78 is on the shelf and how deep the seafloor is in relation to the mentioned 140 m. . And how does one explain the enhanced pAl levels in the absence of enhanced dAl? (discussed later I noticed, maybe move this discussion forward or mention it will be discussed later)*

→ We have clarified which station corresponds to the Newfoundland shelf and the depth at which the seafloor is found. We have added a sentence on the probable cause for not seeing elevated dAl when elevated pAl is found.

Page 13 line 15-19: In contrast, on the Newfoundland shelf and margin (St. 78), no enhanced dAl levels were observed near the seafloor (Fig. S6). However, a large input of pAl was observed (station 78), and pAl concentrations increased from 94.6 nM at a depth of 140 m to 550 nM at the seafloor (377 m) (Gourain et al., this issue). Dissolved Al could be scavenged by resuspended particles; thus showing lower levels where elevated pAl levels were observed.

- *Section 3.4.3 this section is very brief. Perhaps some discussion as to why enhanced dAl has been observed at two hydrothermal locations (one very close by) but not at this location or numerous other active hydrothermal vent sites?*

→ Yes, you are right. This section is brief. The reason behind it is that we do not have any prove or complementary data to argue along this section. Thus, any more words would be just a merely speculation. A more detailed discussion on the possibly hydrothermal source of trace metals is given in Tonnard et al. (2018) and Gourain et al., special issue.

Technical corrections:

- *abstract last sentence of abstract is confusing, had to read it several times, please rephrase (possibly two sentences) for increased clarity*

→ We have split this long sentence into two separate sentences.

Page 1 line 27-30: This study clearly shows that the vertical and lateral distributions of dAl in the North Atlantic differ when compared to other regions of the Atlantic and global ocean. Responsible for these large inter- and intra-basin differences are the large spatial variabilities in the main Al source, atmospheric deposition, and the main Al sink, particle scavenging by biogenic particles.

- *Page 2 line 6 from nanomolar to*

→ Corrected

- *12 into the ocean*

→ Corrected

- *26 want is meant by active scavenging*

→ We have now defined active scavenging.

Page 2 line 20-25: Removal of Al in oceanic waters occurs through particle scavenging with subsequent sinking of the particulate matter (Orlans and Bruland, 1986). This removal occurs via both active and passive scavenging processes. Active scavenging occurs when dAl is incorporated into the atomic structure of opaline diatom frustules, a process which has been demonstrated in laboratory experiments and is also supported by positive correlations between silicic acid ($\text{Si}(\text{OH})_4$) and Al in depth profiles as a result of the sinking and remineralization of diatomous material

- *Page 5 Line 19 rephrase as readers are not all familiar with this test, e.g. 'A Fisher based test to compare vertical profiles as described by Middag et al. (2015) was used to: : : : This test calculates an integrated p-value as an objective metric to determine how far two profiles are consistent between each other within a given depth interval. This test determined: : :..*

→ This point was addressed by referee 1. We have added the suggested sentence.

Page 5 line 5-9: The Fisher's exact test was used for comparison between profiles as both flow injection data sets were measured with replicates (Middag et al., 2015). This test calculates an integrated p-value as an objective metric to determine how consistent two profiles are within a given depth interval. The test determined no significant difference (i-p-value= 0.2-0.3) within analytical uncertainty comparing the two profiles.

- *Page 5 Line 28 mention were this will be explored*

→ We are sorry not to understand the signification of the previous comment.

- *Page 8 line 3-5 citations needed*

→ We have added citations.

Page 7, line 11-15: Biogenic opal production and biogenic particles play an important role in the removal of dAl in the surface ocean as a result of the high particle affinity of dAl (Moran and Moore, 1988b). Removal of dAl by biogenic particles therefore represents a mechanism which reduces dAl and increases pAl concentrations in surface waters (Moran and Moore, 1988a).

- *Line 12 how would scavenging of Al onto other particles explain the increased pAl/dAl ratio in regions were diatoms were dominant?*

→ You are right. It does not. We have changed this sentence in order to make our point more clear.

Page 7 line 24-27: This conclusion is supported by increased pAl to dAl ratios where surface waters were dominated by diatoms (Fig. 5), as possibly a consequence of active dAl incorporation into siliceous shells (Gehlen et al., 2002) or scavenging of dAl onto biogenic opal (Moran and Moore, 1988a).

- *Line 14 what is meant by a 'high correlation'?*
→We wanted to say strong correlation. As such we have changed the word high for strong.
- *Page 9 line 9 here and throughout the ms, refer to transect (GA04N) rather than cruise 64PE370 to be consistent with references to other transects*
→We have changed all 64PE370 for GA04N
- *Page 10 line 1 rephrase e.g. 'had coefficients of determination above $R^2=0.89$ '*
→Changed
- *line 28 'concentrations was' singular plural mismatch*
→Changed
- *Page 11 line 10-11 the ranges in dAl indeed overlap, but the medians are quite different so quoting the medians here is confusing*
→That is right. We have removed the brackets with the median value.
- *Page 12 line 27 elsewhere in the ms salinity is used rather than S (I prefer salinity)*
→We have changed S for salinity throughout the whole ms.
- *Page 13 line 1 core between 1000 and 1200 does not correspond to depth of Al maxima mentioned just previously*
→Indeed, the maximum dAl concentration is found at station one at ca. 900 m depth. Once we follow the transect, the core of maximum concentrations was found between 1000 and 1200 meters depth (neutrally density layer $27.6-27.8 \text{ kg m}^{-3}$). This difference is possible due to a different branch of the MOW flowing northwards as the branch we plot which follows a westward direction. The core depth chosen is based on salinity which shows the highest values at this depth. We have rewritten the sentence.

Page 12, line 15-17: Figure 7 displays dAl & pAl versus salinity in the MOW for the neutrally density surface layer (σ_t) between 27.6 and 27.8 kg m^{-3} , corresponding to a MOW core depth, based on highest salinity values, between 1000 and 1200 m.

- *Line 23 why z (not defined) rather than depth?*
→ We have modified z for depth. We also modified z for depth at page 13 line 28.
- *Fig1a what is the red square on Greenland?*
→ The red square represents the location where fjord and Iceberg samples were taken. This has been added to the caption.
- *Fig3 name the land masses on the map for those not familiar with this region*
→ We have added Iberian Peninsula to the plot.
- *Fig 4 caption is not clear, what is fig3 in the manuscript?*
→ We have modified the caption of figure 4 and clarify the issued mentioned.
- *Fig 5 caption, wouldn't 'around the southern tip of Greenland' be more appropriate than 'around SE and SW Greenland'?*
→ We have modified the caption and modified SE and SW Greenland for around the southern tip of Greenland.
- *Table 1 number of significant figures is inconsistent and seems inappropriate. Uncertainties on the slopes and intercepts should be reported (and hence for the endmember estimation)*
→ We have provided consistency for the significant figures. We have reported the uncertainty for the endmember estimation.
- *Fig 6 b Was this plot was made in ODV? Caption implies this*
→ No. Caption has been changed.
- *Fig 7 number of significant figures does not seem appropriate and why now GA01 rather than GEOVIDE? And why is the neutral density window reported in the legend for GA01 data and not the others, was it different? This figure could be made to look better if made in another programme (same for fig 8).*
→ We have changed the number of significant figures. We also have changed GA01 for GEOVIDE throughout the text. You are right. They are different. We have acknowledged the difference in the caption. For GA03 and GA04N only the highest dAl concentration

between a neutrally density surface layer (σ_{θ}) between 27.6 and 27.8 kg m^{-3} is plotted. We have redone the figure.

- *Fig 8, why not station 78 as shelf station?*

→ We have not added shelf stations because it would have been difficult to interpret the plot. However, we now have included a figure in the SI showing dAl and pAl for station 78 and 2 on the Newfoundland and Iberian shelf, respectively.

Page 12 line 5-7: On the Iberian shelf and margin enhanced dAl concentrations were observed near the seafloor (station 2: up to 21 nM at a depth of 140 m; station 4: up to 27 nM at a depth of 800 m) associated with enhanced pAl concentrations of up to 1.5 μM at station 2 (Fig. S6).

Page 12 line 15-16 In contrast, on the Newfoundland shelf and margin (St. 78), no enhanced dAl levels were observed near the seafloor (Fig. S6).

Referee 3 (Joseph Resing):

P2

- *L7 Orians and Bruland are not the primary reference for hydrolysis of dissolved Al. They do not show this to be true, they reference someone else*

→We have removed Orians and Bruland and just left Roberson and Hem.

- *L17 There are other papers that show a fluvial source of Al. There are two Brown and Bruland papers, and the Moran and Moore paper referenced here also shows a fluvial source. I think another one of your references or associated references shows a fluvial source as well. I suggest that you reference those papers for a fluvial Al source. Such referencing supports your Iberian margin surface Al data from the Tagus estuary.*

→We have added more references for these lines.

Page 2 line 10-14: Fluvial inputs were historically considered a dominant source of Al to the surface oceans (Stoffyn and Mackenzie, 1982). Whilst Al removal during estuarine scavenging processes appears to strongly reduce the riverine Al outflow (Hydes, 1989) a significant fluvial source of Al is still generally observed (Brown and Bruland, 2009; Brown et al., 2010; Grand et al., 2015).

- *L20 "Sediment resuspension processes at ocean margins with strong boundary currents (Jeandel et al., 2011) and in benthic nepheloid layers (BNLs) (Hesse et al., 1999; Middag et al., 2015b; Moran and Moore, 1991) represent important sources of Al to the deep ocean."*

Should be rewritten "Sediment resuspension represents an important source of Al to the deep ocean especially along ocean margins with strong boundary currents (Jeandel et al., 2011) and in areas with benthic nepheloid layers (BNLs) (Middag et al., 2015b; Moran and Moore, 1991).

→We have modified this part as suggested.

- *L27 Probably worth defining active and passive scavenging?*

→We have added a definition for active and passive scavenging.

Page 2, line 20-30: Removal of Al in oceanic waters occurs through particle scavenging with subsequent sinking of this particulate matter (Orians and Bruland, 1986). The removal occurs via both active and passive scavenging. Active scavenging occurs when dAl is actively incorporated into the atomic structure of opaline diatom frustules, a

process which has been demonstrated in laboratory experiments and is also supported by positive correlations between orthosilicic acid ($\text{Si}(\text{OH})_4$) and Al in depth profiles upon the sinking and remineralization of diatomous material (Gehlen et al., 2002; Hydes et al., 1988; Hydes, 1989; Middag et al., 2009; Middag et al., 2015; Moran and Moore, 1988). Passive scavenging is defined as dAl being adsorbed onto any particle surface without being intrinsically incorporated into cellular structures. This is inclusive of adsorption onto biogenic particles. Evidence for the *post-mortem* incorporation (e.g. passive scavenging) of Al onto diatom frustules and concomitant removal from the dissolved phase is given in Koning et al. (2007) and Vrieling et al. (1999).

- *Note that Orians and Bruland say “The markedly different distributions of dissolved Al and the nutrient silicate in the open ocean, as well as their opposite inter-ocean fractionation trends support a passive adsorption removal mechanism.” Unless I don’t understand this paragraph, I don’t think that this is what you are saying.*

→ Yes. You are right. It is clear that in the Pacific Ocean, due to the scavenging nature of dAl versus the recycled nature of Si, both elements are decoupled and do not show a correlation. Dissolved aluminium will continuously be scavenged along the flow path of water masses. Thus, observations in the Pacific would, generally, conclude that passive adsorption is the main mechanism removing dAl. However, in our study region, with younger water masses, stronger phytoplankton blooms (including diatoms and coccolithophorids) and freshly exported organic material it seems that the cycle of dAl is much more closely related to primary productivity than is the case in the Pacific gyres.

- *Also, does this section preclude the removal of Al by particles that are not biogenic Si?*

→ No, not at all.

- *Can Al be scavenged by other particles? Must it be incorporated into the frustule or can it be on the organic matter?*

→ We have rewritten this paragraph in order to make clear that dAl is not necessarily removed by biogenic Si and that it also can be removed by other biogenic particles. See the above answer to comment “L27 Probably worth defining active and passive scavenging?”.

- *L5 do you mean high resolution sections as opposed to profiles?*

→Yes. We have rewritten this sentence.

Page 2, line 31-32: In the North Atlantic (40° N-65° N) vertical dAl profiles combined with high resolution sections were scarce prior to the GEOTRACES era.

- *L8 there must be more N Atlantic data from Hydes, Measures and others that you could reference here?*

→Indeed there are more studies which we could reference. We have tried (sections 3.2.3, 3.3, 3.4.1) to compare our data with the most recent GEOTRACES sections as all the datasets are going through the process of intercalibration.

P4

- NADR is used only once in the text (with two of the letters inverted.)

→Done

- SANARCT does not appear elsewhere in the text according to the search that I did. Please remove the parentheses and delete SANARCT

→It does appear at page 8 line 23. To avoid confusion with the term Arctic we have rename this province as SANA (Subarctic North Atlantic) throughout the manuscript-

- ENACW does not appear elsewhere in the text according to the search that I did. Please remove the parentheses and delete ENACW

→Done

- *L26 change "present low levels of O2" to "is depleted in O2"*

→Changed

P7

- *The surface distribution in Al is hard to understand from the figures. Figure 2 needs to use a split axis so that values between 0-10 nM can be seen. I suppose that a log axis would work, but that makes it hard for the reader to gauge what the actual Al values are. Without split axis this figure is not very useful and the fresh water and Iberian input can be summarized in a couple of sentences.*

L5-12 If we could see the data in the figure then this section would not be so painful to read

→We have changed the plot. We also have added biogenic silica. The shelf stations have been removed from this plot as they are shown in figure 3 and 4.

- *L19 “in line” should be changed to “consistent with”*

→Modified.

P8

- *L7 NADR not NARD*

→Changed

- *L7 ARCT not defined anywhere*

→Indeed, this should be SANA which is defined in Page 5 line 24. We have modified it to SANA.

- *Tonnard et al should be made available. You list an A and a B, but don't list the citations in the bibliography.*

→Tonnard et al., (a) has now been published in biogeosciences discussion. We have added the citations to the bibliography.

- *L10 Figure 6 called before figures 3, 4, and 5.*

→ We have changed the figure numbers.

- *L14 High correlation in euphotic zone between dAl and SiOH4. Why is this not plotted in an x-y plot? Depending on the time of year (pre-bloom, post-bloom, pre-dust, post-dust), this plot could be meaningless. What about correlations with P*Si* and pAl?*

→The sentence was not clear. We made the clarification suggested by referee 1. We meant strong correlation with depth and not in surface waters. We have now added BSi data into figure 2 and commented on this.

Page 7, line 20-29: Elevated bSi and pAl concentrations and low dAl concentrations coincided with diatom dominated phytoplankton communities for the ENAB, IrB, and LB (Fig. 2). In contrast, elevated dAl and low bSi and pAl concentrations coincided with coccolitophore dominated phytoplankton communities for the IcB (Fig. 2). The latter could suggest a preferential scavenging of dAl by diatoms rather than by coccolitophorids. This conclusion is supported by increased pAl to dAl ratios where surface waters were dominated by diatoms (Fig. 5), probably as a consequence of active dAl incorporation into siliceous shells (Gehlen et al., 2002) or scavenging of dAl onto biogenic opal (Moran and Moore, 1988a). One way ANOVA analysis was performed for the pAl to dAl ratio in the surface waters (>50 m) in each of the four basins which showed strong dAl to Si(OH)₄ correlations with depth (LB, IrB, IcB and ENAB, see section 3.3.1).

P9

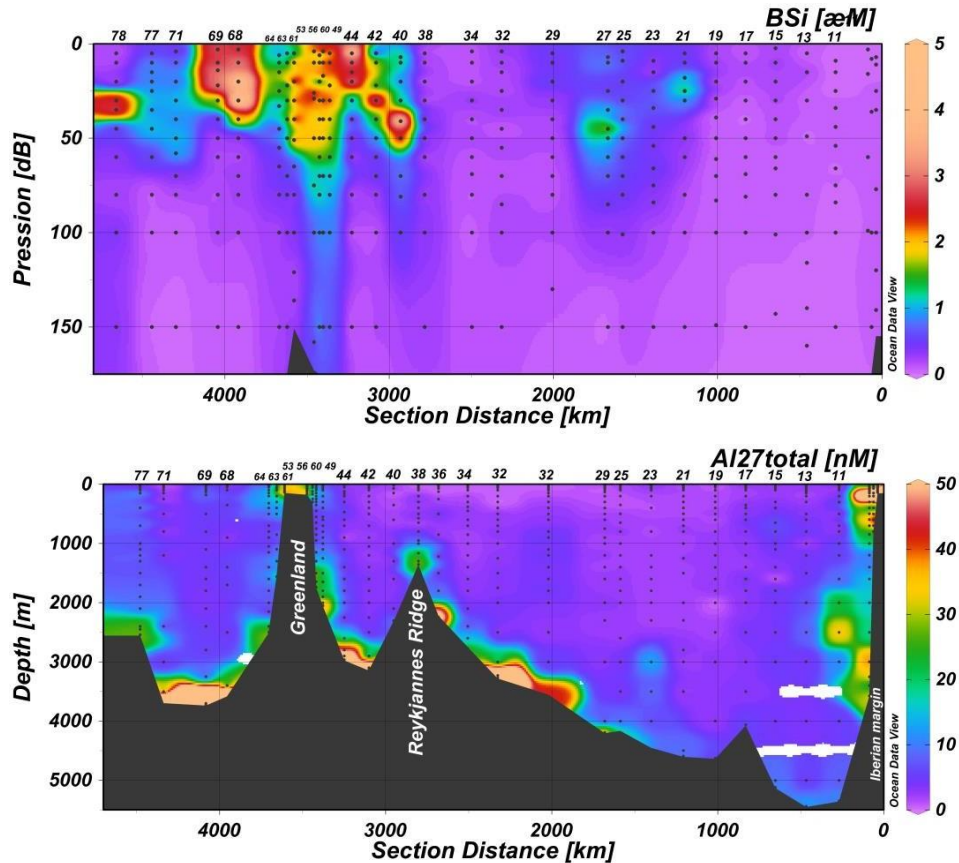
- “we conclude that the observed differences in pAl to dAl ratios were related to diatom abundance.”

→ We have modified the sentence as we are not able to say with 100% of certainty that the differences in pAl to dAl ratios are solely a function of diatom abundance.

Page 8 line 7-10: Considering the low aerosol deposition, reduced fluvial inputs into the tested basins (ENAB, IcB, IrB and LB), and elevated levels of bSi, we conclude that the observed differences in pAl to dAl ratios were mainly related to the of diatom abundance.

- You don't present any biogenic silica data. You don't document that Al is not scavenged by other particles. As a result this appears to be mostly unsupported inference. With a combination of pSi, pCa, pAl and dissolved Al can we make an advance in our understanding of the marine Al cycle? Compared to many studies the combined use of particulate and dissolved chemistry can be a powerful tool.

→ Yes, the combination of dissolved and particulate datasets is one of the goals of large research programmes such as GEOTRACES and will help us to understand from an holistic point of view key questions regarding the Al biogeochemistry. However, it is sometimes impractical to present, interpret and combine all these datasets in one publication. We do not present BSi and pAl in this manuscript as there are going to be published by other authors within the GEOVIDE special issue. pCa data is not available for this section. However, we do have permission from the data owners to show the section plots of pAl and bSi (see below). The pAl and BSi sections are being published in the GEOVIDE special issue by Gourain et al., and Sarthou et al.,.



It is to notice that BSi was analysed from samples collected from the regular CTD and not from the trace metal clean CTD. Thus, the sampling depths do not always coincided between both CTDs.

- Can you say anything about the role of coccolithophores? The LeMaitre paper documents their abundance along your section. If you could discount their role in the scavenging of Al from the surface ocean, that would be a very novel finding. Is there a novel or transformative result here? So far your manuscript repeats what others have found and the results seem mostly as expected. I would think that other classes of particles would scavenge Al, especially large quantities of particles produced during phytoplankton blooms independent of the type of phytoplankton. However, perhaps my thinking is incorrect. Can your data address these questions?

→ We suspect that dAl is scavenged by different types of particle and that the amount of scavenged dAl will increase as a function of the amount of each particle class present. As such, the North Atlantic Ocean represents a key area to investigate this process. However, it can be difficult to draw correct conclusions with just this study as the strong phytoplankton blooms in

the North Atlantic are dominated by diatoms and after diatom decay, by coccolithophorids. It would be really interesting; as such we have added a sentence in the conclusions section, to investigate the distributions of dAl and pAl along with bSi and pCa during the duration of a phytoplankton bloom.

Diatoms, potentially, scavenge more efficiently dAl from surface waters than other phytoplankton types (e.g. coccolithophorids). The reason is that dAl is adsorbed onto diatom frustules (passively) and/or incorporated Al into the structure of frustules (Whether this is an active or passive mechanism remains unclear).

CHEMTAX data are published by Manon Tonnard and co-authors in the special issue. Based on the CHEMTAX data, diatoms were the dominant phytoplankton community in the ENAB (40%), IrB (>60%), and the LB (>60%). In contrary, in the IcB coccolithophorids dominated the phytoplankton community (50%). From the CHEMTAX data it is clear that any attempt to disentangle the relationship that coccolithophorids and diatoms exert on the behaviour of dAl and pAl in surface waters would be challenging as the phytoplankton community composition was, in all the different basins, highly variable.

The enhanced levels of bSi in the SANA region (IrB and LB) indicate the presence of diatoms which is confirmed by the CHEMTAX data. Based on the ANOVA analysis, the ratio of pAl to dAl was significantly different for the IrB and LB compared to the Iceland and East North Atlantic Basins. In contrast, no significant difference in the ratio for pAl to dAl was found between the IcB and ENAB.

Page 11 lines 31-35, page 12 line 1: However, it should be noted that dAl removal by diatom production is not necessarily the only reason for the nutrient type distribution. Whilst we did not observe enhanced pAl concentrations where coccolithophorids were dominant (e.g. IcB; section 3.2.2), we assume that also other biogenic and non-biogenic particles (e.g. CaCO₃, organic carbon, lithogenic particles, zooplankton fecal pellets) will contribute to the vertical export of surface dAl into the deep ocean.

- *L7 should you reference figure 2 here as well?*

→We have also included figure 2.

- *L7-8 “which decreased westwards to 5.6 nM (station 11).” I would consider changing this to “which decreased westwards, reaching 5.6 nM at station 11.”*

→Changed.

- *L16 is there any relevance to the estuary being polluted? The more important point is that rivers can be large sources of dissolved Al (Brown and Bruland papers plus another one that you reference here.*

→ We agree with you. We have removed the term polluted. We have added additional references.

Page 9 line 4-6: Our results indicate that a fraction of riverine dAl can be advected offshore, as observed previously in the Bay of Bengal (Grand et al., 2015), Gulf of Alaska (Brown et al., 2010), and coastal waters of Oregon and Washington (Brown and Bruland, 2009),....

- *P10 L3 “were” not “was”*

→ The discussion in this paragraph has been rewritten according to comments from Rob Middag (Referee report 2).

P11

- *L7 -10 “Increased aerosol deposition was observed during GA03 (Shelley et al., 2015) in comparison with GEOVIDE (Shelley et al., 2017). Thus, enhanced dAl for GA03 could be related to enhanced removal of dAl in surface waters and concomitant remineralization of biogenic particles following the decline of the late summer-autumn bloom.” It seems that if you had more aerosol deposition during GA03 then you would expect higher dAl and remineralization is not required. Why do you make this point? What have I missed?*

→ We have rewritten these sentences for clarification. We argue that the accumulation of dAl in surface waters from atmospheric aerosol deposition during June, July, August, and September and the removal occurring during the bloom period (months) with dAl subsequently released at depth may have been the possible source for the enhanced concentrations observed. However, we still state at the end that this remains speculative.

Page 10 line 24-28: The region is known to receive enhanced atmospheric aerosol deposition (Mahowald et al., 2005). A possible explanation for the difference between GA01 and GA04N with GA03 may be that dAl is accumulated in surface waters during June to September; thus, following the late summer-autumn bloom in the Iberian Basin it is possible that the excess of dAl accumulated in surface waters is removed by particles produced by the bloom and then released in sub-surface waters. However, this explanation remains speculative.

- L8 “in surface waters” should be “from the surface waters”

→ Changed.

- L10 give depth range of “sub-surface”

→ It was given some lines before in the same section. Page 10 line 7.

- Section 3.3.1 Need to mention scavenged type distribution in Pacific. Certainly Orians and Bruland and Brown and Bruland papers think about their data in terms of scavenging and not as nutrient-like. Also may need to take a closer look at the Barrett et al and Measures 2008 in terms of this question (not sure which of these two).

→ We have added that dAl in the Pacific Ocean shows a scavenged type distribution.

Page 11 line 9-10: A scavenged type distribution for dAl has also been described for the Pacific Ocean (Orians and Bruland, 1985).

P12

- L1 : “this vertical distribution coincides” should be “these distributions coincide”

→ Corrected.

- L3 Moore and Millward not in bibliography

→ Corrected.

- L3-5 it is not clear to me that Orians and Bruland nor Middag et al actually showed that dAl was removed onto particle surfaces. Both sets of authors surmised this from their data but did not demonstrate it. Middag et al 2009 summarized other people’s work that made some attempt to look at particulate phases, but Middag et al 2009 did actually not report on particulate chemistry and thus can only surmise that the dAl was moved onto particles. Thus you need to be careful in your references that you actually reference those papers that truly demonstrate your point. Although Middag et al 2009 has a nice summary of some of those papers, he is a secondary and not a primary source.

→ We have rephrased that part.

Page 11 line 15-18: Dissolved Al is considered to be removed from surface waters onto particle surfaces (Moore and Millward, 1984; Orians and Bruland, 1985), including diatom cells (Gehlen et al., 2002) and subsequently released at depth during the recycling of biogenic particles and desorption from non-biogenic particles.

- L7-8 you seem to imply that diatoms are the only surface that scavenges Al. Moran and Moore demonstrate that Diatoms are an important scavenger of dAl but do not address other substrates.

→ We have modified the sentence.

Page 11 line 18-21: In our study region, diatoms dominate the phytoplankton communities at the early stage of the spring bloom (Brown et al., 2003), and are an important producer of biogenic silica (bSiO₂) (Nelson et al., 1995) which is, along with other non-biogenic particles, a major carrier for scavenged Al (Moran and Moore, 1988b; Stoffyn, 1979).

- *Are “Lemaitre et al 2017” and “Lemaitre et al this issue” the same papers? IF so I can’t find the biogenic Silica in that manuscript. The full bSiO₂ data set would inform this manuscript.*

→ No. This was a mistake. They are two different manuscripts. The bSi (in situ pumps) is going to be presented in Lemaitre et al (b). It should be soon available on BGD as part of the special issue. The bSi from ctd casts is being presented by Sarthou and co-authors.

- *L16-17 “suggests that the net remineralization of dAl from particles was larger than the net removal of dAl from scavenging.” This implies that dAl should be building up over time or that as the water moves away new water is continually fortified by new remineralization.*

→ Correct.

- *The idea that remineralization is a dominant process is a key one in understanding Al biogeochemistry and it would be nice to have all of the data to properly demonstrate it. It really seems odd that the particulate Al and pSi are not a part of this manuscript. Are there changes in the ratio of pAl to dAl at different depths?*

→ The ratio between pAl and dAl decreased, generally, with depth. However, close to the seafloor and due to resuspension of sediments the pAl to dAl ratio increased again. This can be observed in Fig. 5 c.

- L27 sentence about figure 7 does not make sense.

→ We have rewritten the sentence and took out the word correlations.

Page 12 lines 15-17: Figure 7 displays dAl & pAl versus salinity in the MOW for the neutral density surface layer (σ_t) between 27.6 and 27.8 kg m⁻³, corresponding to a MOW core depth, based on highest salinity values, between 1000 and 1200 m.

P13

- L14 need a reference for Al in pore waters. In doing this review I came across Stoffyn-Egli 1982, but I would love to know if you are aware of anything else.

→ Added. Another reference is Van Beueskom et al. (1997)

- L19 Sherrell and Boyle do not discuss dissolved Al.

→ We have removed this citation.

P14

- L1. EGC is used 2 times in the text and once in the figures. Thus you should use “East Greenland Current (EGC)” each time. WGC is only once in the text and once in the figures. Also WGC is not defined when it is used in the text.

→ We have added the full name for the abbreviations mentioned.

- L14 might consider using beam attenuation instead.

→ Considered and changed as suggested.

- L20 light transmission is nice, but particulate Al would be much much better. Perhaps the pAl is from pumps and not from the same samples? Still adding pAl would be helpful.

→ A similar issue was raised by Dr. Middag’s review. We have now included a plot in the SI (Fig S6) showing the distribution of dAl and pAl with depth for station 2 and station 78 over the Iberian and Newfoundland shelf, respectively. The pAl data were analysed from samples collected using the trace metal clean CTD.

- L25 do you mean that dissolution/ remineralization from particles rather than partial dissolution of resuspended sediments? Do you mean a dominant process in supplying dAl to the mid and deep ocean?

→No, we meant that scavenging of dAl by resuspended particles may be dominating over Al dissolution from resuspended particles and that this may be the reason why no significant enhanced dAl concentrations are observed when enhanced pAl concentrations are present. We have rewritten this part and added a sentence for clarification

Page 14 line 4-7: These results suggest that, occasionally, scavenging of dAl onto resuspended particles may be a dominant process rather than partial dissolution of Al from resuspended sediments. However, the mechanisms controlling either a net dissolution or scavenging of Al from or by resuspended particles remain unclear.

- L26 should be “general increase in Al”

→Changed.

P15

- L11 use “were” not “was”

→Corrected

- *Figure 1 NAST, NADR, ARCT should be placed within or above boxes instead of letters A, B, and C. All these acronyms are confusing enough, but then adding tertiary relationship makes it very painful to those of us who don't know the region. The reader should be able to look at the figure and understand what is being said. The ideal figure is one in which the reader only needs to read the figure caption once or not at all. It would be good for the basins to be listed as well. For example you list box B as NADR but often talk about the Iceland Basin. It would be helpful for your readers to look at the map and see where the Iceland and other basins are.*

→We have redone the figure.

- *Figure 2. the scale makes it so that the reader cannot see all of the data. Please use a split axis. There is a lot of discussion in the manuscript about these data and I cannot see the features. This is the only way for a reader to assess the meaning (and the quality??) of the data. IF you need to*

make a separate panel, then it is worth it. Need to show station 60 in this figure because it is shown in figure 5.

→ We have redone the figure. We now show all stations except the ones over the Iberian and Greenland shelf. We have included biogenic silica in this plot. The Iberian and Greenland shelf stations are shown in figures 3 and 4.

- *Figure 3. Very nice figure. Is there anyway to stretch the color bar to let us see how dAL at stations 11-17 changes. This can also be accomplished by making the suggested changes to figure 2.*

→ Thanks. It has been changed on figure 2.

- *Fig 6 need label on color bar. A separate section panel covering 0-500m is needed.*

→ Changes done.

- *Fig 6b I can't read any of the fonts on this figure. Increase y-axis font and decrease label interval from 5 to 10. Need to find a way to make x-axis readable as the font is tiny. Inside each of the profile boxes at the top of the figure are labels that could only be read with a zoom. In your figure, going left to right (the way we read) the literature data are in orange, green, and then blue. In your figure caption, these boxes need to be discussed in that order too.*

→ Changes done. Profile boxes are now figure 6c.

- *F6c label needed on color bar with units*

→ Figure 6c is now figure 5. Changes done.

- *Figure 8 Need pAl on this plot or on a parallel panel*

→ We have added a new plot for pAl. Figure S6.